Homogeneous ignition and volatile combustion of single solid fuel particles in air and oxy-fuel conditions

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Abstract

The ignition and volatile combustion of single coal particles were investigated under laminar conditions. Relevant physico-chemical processes were analyzed under conventional and oxy-fuel atmospheres with varying O\textsubscript{2} contents in experiments and simulations. An optically accessible laminar flow reactor with well-defined boundary conditions measured with PIV and quantitative OH-LIF was employed. Multi-parameter optical diagnostics were conducted including OH-LIF, luminescence imaging, and backlight illumination. Simultaneously acquired experimental data allowed for the evaluation of particle size, ignition delay time and volatile combustion duration for individual particles. A statistical analysis revealed the improved accuracy of OH-LIF compared to luminescence imaging regarding ignition detection. Simulations within an Eulerian-Lagrangian framework were introduced and validated against experiments. On this basis, particle temperatures, local gas temperatures, and fuel mass fraction were evaluated providing insights into the devolatilization. Both experimental and numerical results indicated that increasing particle sizes significantly retarded homogeneous ignition and volatile consumption. When increasing the O\textsubscript{2} content, a shorter ignition delay time and volatile combustion duration were observed experimentally, which was more significant for larger particles. High slip velocities accelerated convective transport resulting in an earlier ignition and faster volatile combustion. An atmosphere change from N\textsubscript{2} to CO\textsubscript{2} showed an earlier ignition and increased volatile combustion duration for larger particles, whereas the differences

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were insignificant for small particles. Simulation results suggested that the local heat transfer was improved by CO₂, mainly due to the lower temperature sink close to particles and hence higher volatile release rates. As the initial ambient temperatures were similar, the introduction of CO₂ favored homogeneous ignition and slowed down the volatile consumption.

**Keywords:** Single particle combustion, Ignition and volatile flame, Multi-parameter laser diagnostics, Oxy-fuel combustion, Bituminous coal, Combined experimental-numerical approach

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**Highlights**

- Single particle combustion is investigated in a laminar flow reactor with fully-measured boundary conditions using a combined experimental-numerical approach
- Multi-parameter optical diagnostics provide comprehensive data including the particle velocity, ignition delay time and volatile combustion duration
- Simulations assist to interpret the experimental observations with varying particle sizes, oxygen concentrations, slip velocities, and N₂ replacement by CO₂
- The particle size and gas composition dominate the ignition delay time via affecting the particle heating rate
• The N₂ replacement with CO₂ will not delay the ignition if the global gas temperature is constant.

Nomenclature

Greek letters

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<tr>
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<td>( \Delta t )</td>
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Symbols

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<td>( A_{LU} )</td>
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<td>( b )</td>
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**Abbreviations**

- CPD: chemical percolation devolatilization
- COMS: complementary metal–oxide–semiconductor
- DBI: diffuse-backlight illumination
- DLSF: direct least squares fitting
- DTF: drop tube furnace
- EFR: entrained-flow reactor
1. Introduction

On a global level, coal still is the most frequently used resource for electricity generation with a contribution of 36.4% in 2019 [1]. Among the several proposed technical solutions for reducing carbon dioxide (CO₂) emissions, oxy-fuel combustion is a promising technology for coal-fired power generation. This approach uses a mixture of pure oxygen (O₂) as an oxidizer combined with recirculated flue gas which results in a replacement of nitrogen (N₂) in air to CO₂. Consequently, the flue gas mainly contains CO₂ and water (H₂O) which facilitates CO₂ capture and storage. However, the combustion processes are impacted significantly by introducing CO₂ as the major inert species due to its chemical and thermal physical properties. This requires detailed understanding of the underlying multi-phase and physico-chemical sub-processes to enable the application of oxy-fuel combustion technology. Previous studies have been summarized in several reviews [2, 3, 4, 5, 6, 7] providing insights into fundamentals and industrial applications. In the present work, we focus on the fundamental processes of single particle combustion (SPC) in well-defined generic laminar flow conditions to gain a better understanding of ignition and volatile combustion. In the following, selected experimental and numerical studies on pulverized bituminous coal combustion in single-particle mode with high heating rates are briefly summarized.
A brief overview of experimental studies on single particle combustion

In the literature, drop tube furnaces (DTF) and flat flame burners (e.g. Hencken burners or laminar flow reactors) are suitable configurations to achieve high heating rates in the order of \( \sim 10^5 \) K/s. It has to be noted that experimental approaches for the definition of the ignition delay time and the volatile combustion duration are ambiguous. As differences in results may largely depend on the selected measurement techniques and their uncertainties, a comparison of different studies is only possible to a limited extent.

Timothy et al. performed temperature measurements of the sooting flame fueled with pulverized coals using two-color pyrometry in a laminar flow furnace \([8, 9]\) with temperatures of 1250 K and 1700 K. Based on the temperature-time history and derived particle area, the devolatilization time is approximated by the time at which the particle temperature reaches its maximum. They found that the devolatilization time is shortened for increased oxygen concentrations. Bejarano et al. \([10]\) and Khatami et al. \([11, 12]\) conducted a series of experiments on single particle combustion in DTFs with a wall temperature of up to 1400 K using a three-color pyrometer. The volatile flame was investigated based on the typical two-peak temperature-time profiles associated to bituminous coal particles. They found that particle temperature increases with O\(_2\) enrichment but decreases with increasing amounts of CO\(_2\) in the atmosphere. Besides, soot formation is suppressed at high O\(_2\) mole fractions as well as in CO\(_2\) atmospheres \([12]\). However, the effect of particle size on temperatures of particle and gas flames is minor \([10]\).

A restriction of the pyrometric methods is the lowest measurable temperature of typically around 1200 K. The particle temperature could also be possibly biased by the soot flame luminosity or the wall radiation due to the inherent nature of line-of-sight measurements. As recently developed alternatives, advanced optical imaging measurements provide further information for a thorough understanding of single particle combustion. Molina et al. investigated the ignition and volatile combustion of Pittsburgh high-volatile bituminous (hvbf) coal in an entrained-flow reactor (EFR) with ambient gas temperatures \( T_g \) of \( \sim 1250 \) K \([13]\). Time-averaged CH\(^*\) signals recorded with an intensified CCD camera were used to derive the ignition delay time \( t_{\text{ign}} \) and the volatile combustion duration \( t_{\text{vol}} \). It was concluded that O\(_2\) enrichment significantly reduces \( t_{\text{ign}} \). Further, the presence of CO\(_2\) retards \( t_{\text{ign}} \) of single coal particles, whereas the volatile combustion duration \( t_{\text{vol}} \) is affected marginally. They reasoned that the increase in the volumetric heat capacity (the heat sink, \( \rho c_p \)) of CO\(_2\) leads to retarded gas-phase ignition. Shaddix et al. \([14]\) conducted experiments in similar conditions by imaging the high-temperature sooting flame using an intensified CCD camera. Here, \( t_{\text{ign}} \) and \( t_{\text{vol}} \) are statistically evaluated by classifying
flame images into different combustion stages according to soot shape and intensity. They concluded that increasing oxygen concentration accelerates particle ignition and volatile consumption for both N₂ and CO₂ atmospheres. Moreover, both \( t_{\text{ign}} \) and \( t_{\text{vol}} \) seemed to be greater when N₂ is replaced by CO₂. Khatami et al. [15] performed high-speed imaging in a DTF to detect ignition based on the soot flame luminosity. Their results showed that with increasing O₂ mole fraction, \( t_{\text{ign}} \) decreases significantly in a CO₂-enriched environment, while remaining almost unaffected in an N₂-enriched atmosphere.

However, soot particles are essentially produced in the middle-to-late stage of volatile combustion. Intermediate species, such as OH or CH radicals, have been seen as proper indicators for gas-phase flames and have been utilized to image the reaction zone using advanced laser diagnostics. Köser et al. applied high-speed imaging using planar laser induced fluorescence of the hydroxyl radical (OH-LIF) to visualize the single particle volatile flame in a laminar flat flame burner with gas temperatures of \( \sim 1800 \) K [16, 17]. For the first time, the reaction zone near the particle-gas interface was visualized with high spatial and temporal resolution [16]. An increase of the O₂ concentration reduced the stand-off distance of volatile diffusion flames [17]. With an improved multi-parameter measurement approach for the simultaneous acquisition of particle size and flame topology [18], it was found that an increase of the particle diameter significantly impacts the volatile flame duration. Recently, a three-dimensional visualization using a laser scanning system has been applied to reconstruct the three-dimensional volatile flame topology [19, 20]. Gas-phase ignition was initially observed downstream of the particle, where high temperatures and fuel-lean mixtures exist. Increasing particle diameters resulted in larger stand-off distances of the main reaction zone and hence a larger flame size.

1.2. A brief overview of numerical studies on single particle combustion

In the literature related to numerical studies on pulverized coal combustion, it has been found that the devolatilization process in coal combustion is one of the most challenging parts of direct numerical simulation and requires detailed models to simulate the physico-chemical behavior correctly. A well established model for the devolatilization process is the chemical percolation devolatilization (CPD) model which describes the effect of the molecular structure of coal on the devolatilization process in detail [21]. This devolatilization model couples finite rate chemistry with a detailed mechanism for gas phase reactions.

Recently, numerical simulations for single [22] and multiple interacting particles [23] have been performed employing the CPD model [21] in a fully coupled Eulerian-Lagrangian point-particle numerical framework to investigate a number of
aspects including the sensitivity of ignition delay time to temperature, gas composition, particle size, and particle number density. The results have been validated against a series of experiments in a laminar entrained flow reactor [24]. A double peak profile in the time evolution of the volatile release rate has been observed, which strongly affects the ignition delay time. It has been shown that ignition can occur on the onset of the first or the second peak of the devolatilization rate in different initial gas temperatures and particle sizes. It has also been shown that particle Reynolds number and slip velocity between gas and particles have an impact on ignition delay time and the volatile flame which cannot be neglected. The formation of wake flames due to high slip velocities becomes increasingly relevant for flame interactions with multiple interacting particles [25] and turbulent coal combustion [26]. Switching from conventional to oxy-fuel combustion can affect the combustion behavior due to the different thermal and chemical properties of CO$_2$ compared with N$_2$. For instance, several numerical [27, 28, 29] studies indicate, that replacing N$_2$ by CO$_2$ increases the ignition delay time and the duration of devolatilization.

1.3. Objective of the present work

The present work’s aim is the investigation of relevant physico-chemical effects on pulverized coal combustion on a single particle scale. For this purpose, the ignition and volatile combustion of single bituminous coal particles are examined employing both experimental and numerical methods under well-defined laminar flow conditions. Because consecutively performed measurements of different quantities can only be combined using statistics with the need for an evaluation of the repeatability of the investigated process, multi-parameter measurements are conducted in this study such that relevant quantities are measured simultaneously providing a comprehensive data set with high spatial and temporal resolution. The volatile flame is visualized by using OH-LIF to detect the ignition and using luminescence imaging to determine the temporal end of volatile combustion. Additionally, high-speed backlight illumination addresses particle size, shape and velocity. With well defined boundary conditions, experimental results are analyzed showing the importance of particle size and ambient gas composition. Detailed numerical simulations are conducted and validated against experiments with respect to the ignition delay time and volatile combustion duration. Beyond that, simulations provide further insights into particle temperatures, local gas temperatures, volatile release rates, and fuel mass fractions. Combining experimental and numerical expertise, effects of particle size, oxygen concentration, slip velocity and the introduction of a CO$_2$ atmosphere are discussed to understand the ignition and volatile combustion in the present configuration.
The structure of this work is as follows: in Section 2, experimental configurations and data processing procedures are introduced. Subsequently, numerical models and methods are outlined in Section 3. Experimental results are presented and briefly discussed in Section 4. To better understand the observed phenomena, numerical results are included in the discussion in Section 6. Validations of several cases are presented considering realistic inlet boundary conditions. On this basis, physico-chemical processes during the devolatilization and the importance of different parameters are discussed. Finally, the main outcome of the study is summarized.

2. Experimental methodology

2.1. Laminar flow reactor

Experiments were performed in a laminar flow reactor (LFR) with well-defined boundary conditions at the Technical University of Darmstadt. The configuration was introduced in previous works [17, 18] and is illustrated in Fig. 1(a). The LFR consisted of a $80 \times 80$ mm ceramic honeycomb structure, a fused silica enclosure for optical access and a particle seeding unit with a central injection tube with an inner diameter of 0.8 mm. A premixed laminar flat flame (FF) was stabilized above the burner surface. Both the flat flame and particle carrier gas (Jet) were operated with identical inlet gas mixtures. In this study, four N$_2$ atmospheres (denoted as AIR) with different inlet mixtures of CH$_4$/O$_2$/N$_2$ and three CO$_2$ atmospheres (denoted as OXY) with varying inlet mixtures of CH$_4$/O$_2$/CO$_2$ were investigated. Table 2 lists the oxygen concentration in the post-flame environment, flow rates, equivalence ratio $\Phi$, and adiabatic flame temperatures $T_{ab}$ for each condition. Based on the 1D calculation, the produced water vapor concentration was $\sim 14$ vol% and $\sim 16$ vol% in AIR and OXY conditions, respectively. Cai et al. [30] reported that particle ignition is not affected by a H$_2$O mole fraction up to 20 vol% under comparable conditions. Hence, the influences of water vapor concentrations are considered marginal in this study.

The point of heat-up of individual fuel particles was precisely defined by the particle crossing the flame front of the premixed CH$_4$ flame. The flame position, as illustrated in Fig. 1(c), was previously characterized for each condition with a separate OH-LIF measurement. It should be noted that the flat flame revealed a slightly asymmetry which was mainly caused by the inlet flow and the heat loss to the ceramic surface. For all cases but OXY20, which is shown in Fig. 1(c), this imperfection was hardly noticeable as relatively symmetric flame structures were present. When a bituminous coal particle entered the oxygen-enriched high-temperature environment, it experienced a heating rate in the order of $\sim 10^5$ K/s [18]. This induced a rapid water evaporation followed by the successive release of volatiles, gas-phase ignition,
volatile combustion and char combustion. By modulating the wheel rotation speed of the seeding unit, individual particles were sparsely seeded to guarantee single particle combustion. To give an impression of particle combustion in the LFR, luminous streaks of burning particles are shown in the color image of Fig. 1(b).

Colombian high-volatile bituminous coal particles with two different sievings were used in this study. The samples, denoted as A and B, were sieved to a range of 90 to 125 µm and 160 to 200 µm, respectively. The proximate analysis of the coal composition was 3.5\% moisture(an), 36.9\% volatiles(wf), 54.4\% Cfix(wf) and 8.7\% ash(wf).

Figure 1: (a) Sketch of the Darmstadt laminar flow reactor. (b) Luminosity photograph of burning particles and the flat flame. (c) 2D OH-LIF visualization of the flat flame structure for OXY20 in the region highlighted in (a) by dashed lines.

Table 2: Inlet flow rates defined for different AIR and OXY conditions.

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<td>(\mathrm{CH}_4)</td>
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<td>OXY40 40 vol%</td>
<td>0.29</td>
<td>305</td>
<td>1310</td>
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2.2. Measurements of boundary conditions

2.2.1. Flow field

The laminar flow field was measured using high-speed particle image velocimetry (PIV) as shown in Fig. 2. The carrier gas was seeded with Al$_2$O$_3$ particles (MR-52, Martoxid, $d_{90} = 3 \sim 6 \mu m$) through the particle injection tube. The seeding particles were illuminated by a frequency-doubled Nd:YAG laser (Innoslab, Edgewave) with 2 mJ per pulse at 10 kHz. The laser beam was formed to a sheet with a height of 25 mm and focused at the burner center line to a thickness of $\sim 100 \mu m$. The Mie scattering signals were collected by a CMOS camera (Fastcam SA-X2, Photron) equipped with a macro lens (Sigma, $f = 180 \text{ mm, } f/16$). A band-pass filter (532 $\pm 5 \text{ nm}$) was employed to suppress flame luminosity. The FOV was $20 \times 10 \text{ mm}^2$ (height $\times$ width) with a pixel resolution of 20 $\mu m$.

![PIV Camera](image)

**Figure 2:** Gas velocity field characterization: experimental setup for the high-speed PIV measurements.

The laminar flow field was evaluated using a combined PIV-PTV approach (Davis 10, LaVision) with temporally equidistant images of the seeded flow. The particle vectors were preliminarily evaluated using a PIV algorithm including a multi-pass cross-correlation calculation with gradually decreasing interrogation window sizes. For the subsequent PTV processing, a correlation window size of 16 $\times$ 16 pixel with a vector tolerance of 2 pixels relative to the PIV calculation was applied. The measurements took place at various height positions, such that the region of up to 50 mm above the burner was covered.

2.2.2. Temperature field

Within this study, quantitative OH-PLIF combined with absorption spectroscopy was used to derive a local temperature field. This was possible for lean flames with $\Phi < 0.9$ due to the unambiguous correlation between OH number density and temperature in chemical equilibrium [31]. The correlations have been computed using
0 D reactor simulations for the given atmospheres with varying enthalpy using CAN-TERA [32]. This approach circumvented the usual quantification problems for LIF techniques (e.g. unknown chemical surrounding, detection efficiency). The absorption of laser energy due to resonant energy transfer to the OH molecule was derived by measuring an energy reference before and after the probe volume. The energy ratio for on-resonant measurements was normalized with the off-resonant energy ratio to eliminate influences from non-resonant losses, e.g. on windows and lenses. The integral absorption was calibrated using spectroscopic data from LIFBASE [33] using Eq. 1,

\[
\int [OH](x, y) dr = \frac{A(y)}{g_0 \frac{h \nu_{ij}}{e} B_{ji} D_i f_{B,i}(T)},
\]

where \(A\) denotes the measured integral absorption, \(g_0\) the spectral line overlap integral, \(\nu_{ij}\) the central wavelength, \(B_{ji}\) the Einstein B coefficient, \(D_i\) the degeneracy factor and \(f_{B,i}(T)\) the temperature dependent Boltzmann fraction of the chosen absorption line.

The integral absorption was then distributed along the beam-wise direction \(x\) using the absorption corrected LIF signal \(I_{LIF}^*(x, y)\) itself, see Eq. 2.

\[
[OH](x, y) = \frac{I_{LIF}^*(x, y)}{\int I_{LIF}^*(x, y) dx} \int [OH](x, y) dx.
\]

This approach cancels all influences on the LIF signal that are independent of the coordinate \(x\) for any given height \(y\). In particular, as long as the fluorescence quantum yield is constant along the beam-path, influences from quenching are canceled. This method yields a very high sensitivity and precision, as the equilibrium OH number density doubles approximately every 100 K under these conditions. However, temperature measurements are limited to areas with significant OH content, i.e. \(> 1400\) K. A proper selection of the absorption line is crucial for this approach. This is especially true for atmospheric conditions, where the absorption linewidth is in the same order of magnitude as the laser linewidth. Additionally, the Boltzmann fraction should not vary greatly for the temperature range expected in the investigation. For these reasons, the \(Q_1(6.5)\) transition in the OH \(X^2\Pi \rightarrow A^2\Sigma (v'' = 0 \rightarrow v' = 1)\) at 283.01 nm system was chosen. The Boltzmann fraction for this line varies \(\approx 10\%\) in the temperature range between 1600-1850 K.

The optical setup is schematically depicted in Fig. 3. To excite the OH molecule, a high-speed diode pumped solid state Nd:YAG laser (Innoslab, Edgewave) was used to pump a dye laser (Allegro, Sirah) operated with Rhodamin 6G dissolved in ethanol. The fundamental wavelength and linewidth was measured using a wavemeter.
(WSU-30, HighFinesse) which was calibrated using a temperature stabilized single mode HeNe diode laser. The laser system was operated at 500 Hz repetition rate. OH fluorescence was detected using a high-speed CMOS camera (HSS6, LaVision) coupled with a two-stage image intensifier (HS-IRO, LaVision). A 100 mm lens (2178, Cerco, f/2.8) was employed for high UV-transmission. A narrow-band band-pass filter (T ≥ 90% @ 310 – 320 nm) was used to suppress ambient light as well as Rayleigh scattering. The filter transmits only the (1,1) vibrational band to minimize signal re-absorption [34]. For the absorption measurement, a fraction of the laser beam before and after the probe volume was extracted and guided to a reference cell filled with a diluted Rhodamin 6G ethanol solution. Beam steering effects were minimized by introducing a 4f-imaging lens in the post-burner beam path. Fluorescence was detected using a second high-speed camera (Fastcam SA-X2, Photron) equipped with a 180 mm lens (Sigma, f/5.6).

2.3. Multi-parameter measurements of single particle combustion

Simultaneous 10-kHz multi-parameter optical measurements were performed including planar OH-LIF imaging, diffuse-backlight illumination (DBI) and luminescence (LU) imaging. Figure 4 illustrates the experimental setup including the arrangement of the three diagnostic systems. For the reaction zone visualization using OH-LIF, the dye laser system introduced for the temperature measurements was employed at a repetition rate of 10 kHz. The wavelength was tuned to 283.01 nm to excite the Q1(6.5) line of the A-X(1-0) transition system of the OH radical. The effective pulse energy in the probe volume was approximately 0.3 mJ. The laser beam was formed into a sheet with a thickness of 0.8 mm (FWHM) at the burner center line. The aim of this relatively thick laser sheet was to reduce the probability of out-of-plane loss particles and to allow the signal detection in the early stage of volatile flames. The emission signals were collected by the aforementioned high-speed intensified camera (HSS6 + HS-IRO, LaVision) equipped with a UV-achromatic camera lens.
(Halle, \( f = 150 \text{ mm}, f/2.5 \)). To suppress broadband chemiluminescence and thermal radiation from volatile flames, a band-pass filter \((T \geq 40\% @ 305-340\text{ nm})\) was used and the intensifier gate was shortened to 100 ns. The field of view (FOV) for OH-LIF imaging was \(19 \times 19 \text{ mm}^2\) with a projected pixel size of 25 µm.

For particle shape and size characterization, a DBI system including a CMOS camera (HSS6, LaVision) and a high-power LED (IPS, ILA) was inclined by 17° to the OH-LIF detection system. The LED was operated at 10 kHz with a peak wavelength of 525 nm and a pulse duration of 1 µs. The CMOS camera equipped with a long-distance microscope (SK2, Infinity) and a band-pass filter \((525 \pm 25 \text{ nm})\) detected the particle shadow images with high spatial resolution. With a projected pixel size of 10 µm, the FOV of \(13 \times 5 \text{ mm}^2\) (height \( \times \) width) covered the region from the LFR surface to 13 mm along the \(y\)-axis.

To track the temporal evolution of burning particles, another CMOS camera (Fastcam SA-X2, Photron) combined with a macro lens (Sigma, \( f = 180 \text{ mm}, f/5.6 \)) was employed to image the luminous flame. A band-pass filter \((380-492\text{ nm}, \text{Semrock})\) was used and the recorded intensity was dominated by the thermal radiation of soot, tar and particle surface [18]. The FOV of \(65 \times 10\text{ mm}\) (height \( \times \) width) allowed a particle tracking throughout the entire volatile combustion stage. Due to the large field of view, the projected pixel resolution was restricted to 60 µm but was still sufficient to resolve volatile flames with a size of a few millimeters.

![Figure 4: Experimental setup of simultaneous multi-parameter measurements using OH-PLIF, LU and DBI.](image)

### 2.4. Multi-parameter data processing

The utilization of multi-parameter measurements is essential to obtain case-specific information about ignition and volatile combustion. This is important as the measured quantities of interest are directly linked to each other, enabling a thorough statistical analysis with reduced complexity. Correct data interpretation and

14
better understanding of the underlying sub-processes rely on appropriate data processing strategies. Several crucial parameters relevant to single particle combustion are derived and subsequently, corresponding processing procedures are described in detail.

As one of the key quantities, the ignition delay time $t_{\text{ign}}$ is determined by temporally and spatially tracking the OH-LIF signal. Instantaneous laser profiles are obtained from the background OH-LIF intensity associated with the post-flame region, where a homogeneous distribution of OH-radicals from water dissociation is assumed. The OH-LIF images are shot-for-shot corrected in terms of the pulse energy fluctuation and the sheet inhomogeneity. The signal-to-noise ratio (SNR) is estimated to be approximately 12 and the maximum signal-to-background ratio is around 10. The signals are normalized based on the background intensity and further denoised by using a $5 \times 5$ Gaussian filter. The particle centroids are extracted from simultaneous DBI recordings and used as position references for particle tracking. Only the particles with a sufficient distance of $\geq 4\, \text{mm}$ to the nearest particle were included in the data analysis which guaranteed single-particle combustion mode. As shown in Fig. 5 (a,c,d), a $2 \times 2\, \text{mm}^2$ region of interest (ROI, dashed line) is defined centered around the particle centroid (red cross) in the pre-processed OH-LIF images. By evaluating the background intensity, an intensity threshold of 1.2 is applied for binarization in Fig. 5 (b,d,e). Within the ROIs, a signal and structure (SAS) analysis was performed. The ignition time is determined to when (1) the mean OH-LIF intensity increases over the threshold and (2) the connected area of the largest binary structure exceeds $1 \times 1\, \text{mm}^2$. The latter criterion is defined based on an estimation of the spatial resolution restricted to the laser sheet thickness. Figure 5 illustratively shows three instants at $t_{\text{ign}} - 0.5\, \text{ms}$, $t_{\text{ign}}$ and $t_{\text{ign}} + 0.5\, \text{ms}$ for a single-particle ignition event. The heating start time is set to $t = 0$ and is defined at the instant at which a particle crosses the flame front of the premixed CH$_4$ flame.

The volatile flame duration $t_{\text{vol}}$ is determined by combining $t_{\text{ign}}$ from OH-LIF measurements and the end time of volatile combustion $t_{\text{vol,end}}$ from luminescence measurements. The particle positions extracted from DBI images are used to initialize the temporal tracking of LU signals. The area of LU signals $A_{\text{LU}}$ is evaluated by defining an intensity threshold which is placed marginally (i.e. $\sim 10\%$) above the background level. Figure 6(a) shows the temporal variation of the LU signal area of a single particle flame. Four individual instants are highlighted and visualized in Fig. 6(b-e). The color scale has been adapted to match the low intensity level. A few milliseconds after the onset of ignition, the size of the gas flame increases and then decreases to a local minimum. This implies a transition from the formation of a diffusion flame to the completed fuel consumption via gas-phase oxidation [18].
the subsequent instants, no significant increase in $A_{LU}$ is noticeable since luminous signals are dominated by particle surface reactions. However, the luminosity intensity increases again (not shown) due to particle surface reactions. Hence, the local minimum area is used for an appropriate determination of the end time of volatile combustion $t_{vol, end}$. In a few cases, the luminosity intensity disappears at the end of volatile combustion and appears after char combustion begins. The $t_{vol, end}$ is then determined at the time point of the first disappearance of luminosity signals. Based on this, the pre-ignition, volatile combustion and char combustion stages are temporally classified for each single particle, as shown in Fig. 6. Consequently, the entire volatile combustion duration is defined as $t_{vol} = t_{vol, end} - t_{ign}$.

The 2D projected particle size and shape are characterized with an in-situ DBI measurement. Figure 7 shows three representative particles with shape parameters given in the table. The boundary detection based on the steep gradient is performed using an adaptive thresholding method proposed in previous work [35, 36].
Figure 6: (a) Temporal variation of luminescence signal area $A_{LU}$ of a single particle. (b-e) Four individual 2D visualization of luminescence intensity at successive time steps indicated in (a).

circle-equivalent particle diameter $d_p$ is evaluated based on the enclosed particle area $A_p$. The particle centroids are evaluated and further utilized for particle tracking in simultaneous OH-LIF and LU measurements as discussed before. The particle shape is approximated by an ellipse employing direct least squares fitting (DLSF). Using the major and minor axis, denoted as $2a$ and $2b$ respectively, the aspect ratio is defined as $\beta = a/b$. The error of the area estimation by using the ellipse fitting algorithm is indicated by $\epsilon = (|A_p - A_e|)/A_p$, where $A_e$ is the area of the ellipse. Comparing the three particles in Figure 7, particle P1 and P2 have a similar shape ($\beta$), whereas particle P2 and P3 have similar size ($d_p$); this is in accordance with observations. Hence, it can be concluded that the selected parameters are appropriate for characterization of particle shape and size in this study.

3. Numerical framework and modeling

In the present work, pulverized coal combustion is modeled in an Eulerian-Lagrangian framework, using an Eulerian formulation for the gas phase and a Lagrangian formulation with point particle approximation for the solid particles, which
are fully coupled using a two-way coupling approach. The models were described in detail by Farazi et al. [22, 23]. The governing equations in the gas phase are similar to those applied by Sirignano [37], Attili et al. [38], and Bai et al. [39] with a low Mach number assumption of the Navier-Stokes equations and obeying the ideal gas law for the gas mixture. Chemical reactions in the gas phase are modeled with a finite rate chemistry model using the ITV oxyflame mechanism for methane with 68 species and 906 reactions [40]. Gas-phase radiation is included using the optically-thin approximation. The Eulerian governing equations are solved by using a semi-implicit finite difference code with second-order accuracy in space and time [41] and the Poisson equation is solved applying the multi-grid solver of HYPRE. The Crank-Nicolson method is applied for time advancement along with an iterative predictor-corrector scheme [42].

Coal particles are modeled by the Lagrangian framework, and the equations for determining mass, trajectory, velocity, and temperature of the particles are similar to those applied by Farazi et al. [23]. To describe the devolatilization process, the chemical percolation devolatilization (CPD) model is applied. This model determines the devolatilization rate and composition of tar and light gases based on bond breaking in the molecular structure of reference coals as a function of time according to the study by Grant et al. [21]. In the present work, the light gases consist of CH$_4$, CO$_2$, CO, H$_2$O, and other gases. Other gases are assumed to be C$_2$H$_2$, similar to the assumption by Jimenez and Gonzalo-Tirado [29]. The rate of tar release, which is

<table>
<thead>
<tr>
<th>$d_p$ ($\mu m$)</th>
<th>$\beta$</th>
<th>$\epsilon$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.21</td>
<td>1.18</td>
<td>2.1%</td>
</tr>
<tr>
<td>1.60</td>
<td>1.19</td>
<td>3.5%</td>
</tr>
<tr>
<td>1.61</td>
<td>3.39</td>
<td>1.0%</td>
</tr>
</tbody>
</table>

\[ A_p = \frac{1}{4} \pi d_p^2 \]
\[ A_e = \pi a b \]
\[ \beta = a/b \]
\[ \epsilon = \frac{A_p - A_e}{A_p} \]
also computed by the CPD model, is assumed to be only for \( \text{C}_2\text{H}_2 \). This assumption is also used by Goshayeshi and Sutherland [43] and Tufano et al. [28]. To assess the effect of this assumption on the ignition delay time for the investigated coal and setup, other species like \( \text{C}_6\text{H}_6 \) were used as tar and only a marginal 3% difference in ignition delay time has been observed [22]. Although the ignition delay time of \( \text{C}_2\text{H}_2 \) in a purely gas phase setting is much lower than that of \( \text{C}_6\text{H}_6 \), the marginal difference in \( t_{\text{ign}} \) for the particle setting shows that ignition time is more dominated by the characteristic time required for particle heating. Uncertainties regarding the choice of tar species are taken into account for calculating the ignition delay time.

The coupling between the gas phase and the solid phase has been done through the source terms appearing in the conservation equations by means of a distribution coefficient \( \phi_k \) for each particle, which is computed by a Gaussian function with characteristic width \( L_d \). In this approach, similar to the one used by Farazi et al. [22, 23], the distribution length \( L_d \) is set to \( 2d_p \) where \( d_p \) corresponds to the particle diameter. It has been shown that extending \( L_d \) from \( d_p \) to \( 5d_p \) (corresponding to a volume of \( 125d_p^3 \)) changes the ignition delay time by less than 10%. With the larger values of \( L_d \), the source term is distributed far beyond the reaction zone around each particle, which leads to a significant influence on the ignition delay time [22]. In the Eulerian-Lagrangian approach, the particle equations are derived according to the film model, assuming a uniform gas field around the particle. The domain size is discretized using a uniform grid with cubic cells of length \( d_x \), which is equal to the particle diameter. A convergence study by Farazi et al. [22] showed that for \( d_x < d_p \), the ignition delay time does not change, and for \( d_x > d_p \), the homogeneous combustion cannot be captured around single particles and would lead to a very dilute mixture which could prevent ignition. As a result, \( d_x = d_p \) is used for all simulations. Also, since using the gas phase quantities from grid cells with the same size as the particle might not be consistent with the film model assumption, a filter is applied to provide a smoother field in the gas phase to evaluate the state of the gas surrounding the particle consistently with the film model. The sensitivity studies by Farazi et al. [22] showed that the filter length does not have a significant effect on the results. The accuracy of the models and the methods has been validated against experimentally measured ignition delay times by Liu et al. [24].

For the conducted numerical simulations, an inlet-outlet configuration based on the experimental setup of the flat flame burner is considered. Particles are injected into a hot gas-mixture stream with thermodynamical conditions obtained from the fully burned composition and temperature behind an unstretched premixed laminar methane flame. The initial velocity of the gas stream and particle, temperature, size, and composition are prescribed using experimental data and evolve due to
the particle-gas interaction. In the present work, the flat flame region is neglected because it is assumed to have a negligible effect on the particle temperature and ignition because of the small thickness of the flame and, as a result, the residence time of the particle inside the flat flame is small compared to the ignition time.

4. Experimental results

4.1. Particle size and shape

Figure 8 shows the probability density functions (PDF) of the particle diameter $d_p$ (a), the aspect ratio $\beta$ (b) and the error of the DLSF approximation $\epsilon$ (c) for investigated samples A and B. Approximately 1000 particles from different atmospheres are included for each distribution. To avoid particle swelling and beam steering, only the first appearance of particles after crossing the flame front, which stabilizes approximately at $y = 2\,\text{mm}$, is considered in this analysis. Figure 8(a) shows that both samples deviate from the sieving specifications in the sense that the size distributions are broader and shifted towards larger diameters as coal particles are not perfectly spherical and therefore sometimes pass the sieve although having a larger circle-equivalent diameter than the sieving mesh width. The mean particle diameters (dashed line) are $120\,\mu\text{m}$ and $205\,\mu\text{m}$ for A and B respectively. For $\beta$ and $\epsilon$, no significant discrepancy between differently sieved particles is noticeable. The highest probability of $\beta$ is observed for a value of 1.5 and most particles have an aspect ratio of $\beta \leq 3$. The DLSF particle approximation stays mostly within an acceptable error margin of 5%.

4.2. Gas Temperatures

The mean gas temperature and standard deviation (bar) along the burner center line are shown in Fig. 9 for (a) AIR and (b) OXY conditions. The results are computed for discrete positions along $y$-axis with an equidistant subdivision with a width of 5 mm. The initial temperatures at $y = 2.5\,\text{mm}$ are in good accordance with the adiabatic flame temperatures evaluated by a 1D simulation. Crossing the main reaction zone of the premixed CH$_4$ flame, a noticeable decrease in the slope of $T_g$ at $y = 7.5\,\text{mm}$ is observed for all conditions. Downstream of this, $T_g$ decreases linearly with increasing $y$ with a slope of $\sim 2\,\text{K/mm}$. Since the differences in the flow rate of CH$_4$ are minor, as shown in Table 2, thermal energy produced by premixed combustion is expected to be similar for different conditions. Due to the higher heat capacity of CO$_2$, the entire flow rates in OXY conditions are reduced in order to increase the gas temperature. As one can see in Fig. 9, the temperature profiles along the center line are similar for all investigated atmospheres, which enables reasonable comparisons of single particle combustion.
Figure 8: PDF distributions of the particle diameter \( d_p \), the aspect ratio \( \beta \) and the error of the DLSF approximation \( \epsilon \).

Figure 9: Gas temperature \( T_g \) over the height \( y \) along the burner center line in (a) AIR and (b) OXY conditions. (Outliers at \( y = 17.5 \) mm in AIR30 and at \( y = 32.5 \) mm in AIR20 due to failures/errors of the absorption measurements.)

4.3. Gas and particle dynamics

Figure 10 shows the axial gas velocity \( U_g \) along the vertical coordinate of the burner for (a) AIR and (b) OXY atmospheres. The bars indicate one standard deviation at each \( y \) position. A steep gas acceleration is observed within the first 2 mm, which spatially correlates to the pre-heat zone and main reaction zone of the premixed
CH$_4$ flame. As a result, the gas accelerates due to the steep temperature gradient and thermal expansion across the flame. Since the seeding particles are not uniform in size and hence show a deviating following behavior within the gas flow, a relatively high standard deviation of the measured velocity is observed within the flame. More importantly, high gas velocity gradients exist in the flame region which mainly result in a larger standard deviation. From approximately $y = 3$ mm, the velocity profile starts to stabilize and then remains relatively constant. Comparing different atmospheres, the stabilized gas velocity of AIR conditions is approximately 1.6 m/s and greater than the velocity of 1.3 m/s for OXY conditions. This is attributed to the lower overall flow rates of OXY compared to AIR conditions, as listed in Table 2, in order to obtain similar gas temperature profiles. AIR conditions with different O$_2$ concentrations reveal similar velocity profiles; the same is observed for all OXY conditions as well. A slight discrepancy is noticed for OXY20 at $y = 2$ mm. This is a result of the unevenness of the flat flame structure which is slightly lifted in this condition, as implied in Fig. 1(c). Therefore, seeding particle trajectories might be disturbed by the semi-spherical flame shape close to the particle jet.

![Figure 10: Axial gas velocity $U_g$ over the height along the burner center line $y$ for (a) AIR and (b) OXY conditions.](image)

The particle velocity is evaluated using the temporal evolution of the particle position obtained from DBI measurements. The axial velocity component $U_p$ is calculated with the first derivative of the $y$ component of the particle position using its five-point stencil. Figure 11 shows the statistical results of both particle samples under AIR (a) and OXY (b) conditions. By subdividing the $y$-axis into segments with a width of 1 mm each, it is possible to compute mean values of samples included in the respective segment with standard deviations indicated by bars at each position.
along $y$. As Figure 11 illustrates, the smaller particles of sample A show a higher initial velocity after crossing the flame front due to their lower mass inertia compared to the larger particles of sample B. Afterwards, the velocity slope of both particle sizes is similar, however, smaller particle have a higher acceleration considering the shorter residence time compared with larger particles. This points to the fact that the particle velocity history is essentially dominated by the steep gas velocity gradient within the flat flame, while the aerodynamic drag downstream is of secondary importance.

![Figure 11](image_url)

Figure 11: Particle velocity $U_p$ over the height $y$ in (a) AIR and (b) OXY conditions for differently sized particles A (mean $d_p = 120 \, \mu\text{m}$) and B (mean $d_p = 205 \, \mu\text{m}$).

With knowledge of the mean gas and particle velocities, the mean slip velocity $U_s$ can be calculated and is depicted in Fig. 12 for all atmospheres and both size samples. $U_s$ first increases over the CH$_4$ flame and then decreases with the stabilization of the gas velocity after $y = 3$ mm. The particle Reynolds number can be estimated using the slip velocity and the particle diameter to $Re_p = U_s d_p / \nu$, where $\nu$ denotes the kinematic viscosity of the gas mixture. Based on the gas composition and the adiabatic flame temperature, $\nu$ is estimated to be $\sim 3.3 \times 10^{-4} \, \text{mm}^2/\text{s}$ for AIR conditions and $\sim 2.7 \times 10^{-4} \, \text{mm}^2/\text{s}$ for OXY conditions. Using the maximum slip velocity and mean particle diameter, the particle Reynolds number of small particles is estimated to be 0.29 in AIR and 0.17 in OXY conditions; the particle Reynolds number of large particles is 0.73 in AIR and 0.60 in OXY conditions.

4.4. Comparison of methods for ignition detection

Experimental results of ignition delay times always depend on the uncertainties of both the selected measurement techniques and data processing methods. Köser et
Figure 12: Slip velocity $U_s$ over height $y$ in (a) AIR and (b) OXY conditions for differently sized particles A (mean $d_p = 120 \mu m$) and B (mean $d_p = 205 \mu m$).

al. compared simultaneous OH-LIF and luminescence images at the onset of particle ignition and concluded that the luminescence technique overestimated the ignition delay time due to a strong dependence of signal intensities on the composition of the released volatile matter [18]. This observation is consistent with the results in this study, as discussed in Section 2.4. The temporal difference between the first appearance of luminescence and the ignition delay time determined by OH-LIF signals is quantified by $\Delta t = t_{I,LU} - t_{\text{ign}}$. Figure 13 shows PDFs of $\Delta t$ for all investigated atmospheres with approximately 200 particles included in each case. The PDFs reveal similar trends for all atmospheres with peak values between 3–5 ms. No significant correlation between $\Delta t$ and the $O_2$ concentration can be identified. This can be explained by the large variation of released volatile matter and soot formation of individual particles, which dominate the luminescence intensity. However, a greater $\Delta t$ is observed for large particles which is probably due to the lower particle heating rate and slower devolatilization process (not shown here). The CH* and OH* signals are too weak for time-resolved particle tracking, even with an intensified camera [13]. Broad-band luminescence imaging (without intensifiers) provides higher and therefore more easily detectable signal intensities, whereas the ignition delay time is overestimated, since the luminescence intensity is only strong at high temperatures which are not present at the onset of ignition. This observation needs to be carefully considered to avoid a misinterpretation of experimental data. In this work, high sensitivity and accuracy for the ignition detection are enabled from the time-resolved OH-LIF imaging.
Figure 13: Comparison between LU imaging and OH-LIF with respect to the determination of the ignition delay time. $\Delta t = t_{i,LU} - t_{ign}$, where $t_{i,LU}$ corresponds to the first appearance of the detectable luminescence signal.

4.5. Ignition delay time

The ignition delay time $t_{ign}$ is evaluated for two coal samples for different oxygen mole fractions in (a) AIR and (b) OXY conditions, as shown in Fig. 14. The time at which the particle crosses the flame front of the premixed CH$_4$ flame is set to $t = 0$ and characterizes the beginning of the particle heating. For each condition, more than 60 individual particles are used to determine mean values (dots) and standard deviations (bars). In general, particles with large $d_p$ show a delayed ignition compared to smaller particles. With an increase in the O$_2$ mole fraction within the atmosphere, all particles show a decreasing ignition delay time. Further, the O$_2$ effect seems to be more significant for larger particle sizes. Comparing N$_2$ with CO$_2$ atmospheres, $t_{ign}$ remains in a similar range when considering the same oxygen concentration. For larger particles, however, a slightly lower ignition delay time is observed in CO$_2$ atmospheres. The homogeneous ignition strongly depends on the gas temperature $T_g$ and fuel mass fraction $Y_{F\_0}$ of the volatile-oxidizer mixture in the vicinity of the particle. In the gas phase downstream of the particle, ignition occurs at the lean side of mixtures with increasing temperature; this has been discussed based on the temporal evolution of volumetric volatile flame structures in [20] and in other studies [44]. In the present configuration, the global gas temperatures are similar for all investigated atmospheres. However, the local gas temperature in the vicinity of particles, dominating the particle heating rate and hence the ignition, is strongly influenced by the gas composition and particle size and probably deviates from the global temperatures. A detailed discussion including a numerical analysis is provided.
in Section 6.

Figure 14: The ignition delay time $t_{\text{ign}}$ in (a) AIR and (b) OXY atmospheres for differently sized particles A (mean $d_p = 120\, \mu m$) and B (mean $d_p = 205\, \mu m$).

4.6. Volatile flame duration

Figure 15 depicts the statistical results of the entire volatile flame duration $t_{\text{vol}}$ computed from combined data from OH-LIF and LU imaging. The same data set used for the evaluation of $t_{\text{ign}}$ is applied in this case. With an increase in particle size from 120 to 205 $\mu m$, $t_{\text{vol}}$ rises by a factor of 2 $\sim$ 3. With more oxygen added into the atmosphere, $t_{\text{vol}}$, especially for larger particles, evidently decreases in both $N_2$ and $CO_2$ atmospheres. This indicates that the consumption rate of volatile matter is accelerated by the presence of increased amounts of oxygen. When replacing $N_2$ with $CO_2$, volatile combustion is retarded. Again, this effect is more remarkable for larger particles, while the differences for smaller particles are minor. After the onset of ignition, a gas flame rapidly spreads around the particle, while the particle continues to release volatile matter such as light hydrocarbons and tars. The volatile flame is usually considered as a typical diffusion flame in which the fuel consumption rate is controlled by molecular and thermal diffusion processes. Taking the complexity of this multi-phase phenomenon into account, effects of various parameters are hereafter discussed regarding different underlying sub-processes.

5. Numerical results

For the appropriate interpretation of experimental observations, detailed numerical simulations were performed using a combined Eulerian-Lagrangian framework,
Figure 15: The volatile combustion duration $t_{vol}$ in (a) AIR and (b) OXY atmospheres for differently sized particles A (mean $d_p = 120$ µm) and B (mean $d_p = 205$ µm).

as described in Section 3. The simulations provide access to essential quantities during volatile combustion not readily measurable with state-of-the-art laser diagnostics. Several cases were considered for simulation and are listed in Table 3 denoted by $C_1$-$C_8$. For a direct comparison with experiments, cases $C_1$-$C_4$ employ similar inlet conditions to the experiments and the initial $U_s$ corresponds to the value after particles cross the flat flame at $y \approx 2$ mm. To study the effect of the particle size, $C_5$ and $C_6$ utilize different particle diameters as supplements to the standard size of samples A and B. The slip velocities are linearly interpolated with the assumption that the particle acceleration through the premixed CH$_4$ flame is only a function of the particle diameter. In $C_7$ and $C_8$, $U_s$ is increased to the value of $C_1$ and $C_2$, respectively, which enables a study of heat convection with variations in flow dynamics. For all cases, the inlet gas composition, exhaust gas temperature and velocity remain identical with the experiments, whereas $U_s$ varies only by changing the initial particle velocity.

Table 3: Inlet parameters for all cases investigated in simulation.

<table>
<thead>
<tr>
<th>Case</th>
<th>$C_1$</th>
<th>$C_2$</th>
<th>$C_3$</th>
<th>$C_4$</th>
<th>$C_5$</th>
<th>$C_6$</th>
<th>$C_7$</th>
<th>$C_8$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Atmosphere</td>
<td>AIR30</td>
<td></td>
<td>OXY30</td>
<td></td>
<td></td>
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<td></td>
<td></td>
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<tr>
<td>$d_p$ [µm]</td>
<td>120</td>
<td>205</td>
<td>120</td>
<td>205</td>
<td>80</td>
<td>160</td>
<td>120</td>
<td>205</td>
</tr>
<tr>
<td>$U_s$ [m/s]</td>
<td>0.81</td>
<td>1.17</td>
<td>0.39</td>
<td>0.79</td>
<td>0.2</td>
<td>0.6</td>
<td>0.81</td>
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The numerical results of the ignition delay time and the volatile combustion duration are validated against the available measurements. In the simulation, $t_{ign}$ has
been quantified by taking the time at which 5% of the first peak in OH mass fraction, $Y_{OH}$, has been reached. This definition aims to reproduce the ignition detection of the experimental data, in which the first obvious OH signals above the background is detected. An example of the temporal change of $Y_{OH}$ is shown in Fig. 16. This method is considered to provide accurate values to imitate the OH-LIF measurements.

![Figure 16: Definition of the ignition delay time $t_{ign}$ and the volatile combustion duration $t_{vol}$ in the simulations.](image)

In Fig. 17(a), $t_{ign}$ is compared for different particle diameters $d_p$. In general, the trends of delayed ignition with growing particle diameter are properly captured in accordance with experimental results. For particles with $d_p = 120\, \mu m$, the simulations are able to predict the correct $t_{ign}$ for AIR30, while slightly over-estimating $t_{ign}$ for OXY30. However, considering the fluctuations in the experimental data, numerical predictions remain within the error bars of experiments. Hence, it can be concluded, the simulations provide reasonable predictions in ignition delay time for smaller particles.

For larger particles, an over-prediction of the ignition delay time in the model is observed. Explanations for this deviation can be found in both measurements and simulations. In simulations, the over-prediction can be explained by Eulerian-Lagrangian modeling and the point particle approximation [44]. In the point particle approximation model, coal particles are assumed as points which represent homogeneous spheres without any temperature gradient inside the particle which release volatile matter homogeneously in all directions. Defined by the CPD model, the devolatilization occurs as the bonds in the internal structure of the coal particle break when exposed to heat at a certain particle temperature $T_p$ which is essentially influenced by the initial particle mass and the heating rate. At a high heating rate in the order of $10^5\, K/s$ in the present work, the devolatilization has been experimentally observed as a surface phenomenon [45]. The Fourier number $Fo$ of $\sim 0.08$ and $\sim 0.02$ are estimated for smaller and larger particles, respectively, [20] which implies the existence of evident temperature gradients in the interior of particles. This
Figure 17: Comparison between numerical and experimental results with respect to $t_{\text{ign}}$ and $t_{\text{vol}}$ in AIR30 and OXY30 conditions, including cases from C$_1$ to C$_6$.

leads to a volatile release at elevated particle surface temperatures while the particle core is colder. When assuming a uniform particle temperature, simulations underestimate the surface temperature and thus overpredict the time of volatile release and ignition. As expected, the deviation between experiments and simulations is more distinct for larger particles, shown in Fig. 17a, probably due to the higher temperature gradient indicated by the Fourier number. The point particle approximation is considered as the major reason for the over-prediction of the ignition delay time in the present configuration with high heating rates. To improve the predictions, the temperature gradients need to be numerically considered which requires a particle-resolved simulation in future work. Additionally, in the Eulerian-Lagrangian modeling, the computational mesh resolution is limited by the particle diameter, and the steep temperature gradients in the flow cannot be accurately predicted, especially for larger particle diameters. In experiments, the determination of the ignition delay time with non-quantitative measurements is restricted by the non-linear response of OH signals to the mole fraction. Although the temperature variation is considered as negligible (see Section 4.2), interfering effects of collisional quenching and non-linearity of the image intensifier are not considered.

Additionally, the volatile combustion duration $t_{\text{vol}}$ is validated against experimental data in Fig. 17(b). $t_{\text{vol}}$ is defined in the simulation by the time difference between
the ignition delay time and the time at which \( Y_{\text{CH}} \) decreases to 25\% of its maximum value, shown in Fig. 16. This definition resembles the \( \text{CH}^* \) chemiluminescence measurements in a previous study [13] which is considered to provide accurate values to imitate LU measurements. The CH mass fraction can be represented by a second-order polynomial (black dashed line in Fig. 16) centered at the maximum peak as described in [13] and [24]. From the intersection of the second-order polynomial fit with the baseline, the end of volatile combustion can be estimated. Overall good agreement for the volatile combustion duration can be found. Considering the uncertainties in both simulations and experiments, it can be concluded that the selected model can predict the physical behavior of ignition and volatile combustion of single hvb coal particles reasonably.

6. Discussion

In coal combustion, the energy conservation of coal particles can be described as a change of the particle internal energy by convective and radiative heat transfer. Hence, the particle heating rate can be considered using the following equation (adapted from [5])

\[
\frac{dT_p}{dt} = \frac{6}{\rho_p c_p} \left( h(T_g - T_p) - \sigma(\varepsilon_g T_g^4 - \varepsilon_p T_p^4) \right),
\]

where \( T_p, \rho_p, c_p \) and \( \varepsilon_p \) denote the temperature, density, heat capacity and emissivity of particles in order; \( h \) is the convection coefficient of heat transfer; \( T_g \) and \( \varepsilon_g \) are the temperature and emissivity of gases, respectively, and \( \sigma \) is the Stefan-Boltzmann constant. The wall radiation and the heat release of solid-phase devolatilization reactions are considered negligible under the present conditions. The heating rate is strongly impacted by the particle size, thermal convection, and radiation near the solid-gas interface. In the pre-ignition stage (i.e. water evaporation and particle heating), \( T_g \) is equal to the local gas temperature close to the particle initialized by given boundary conditions, whereas increasing towards the adiabatic flame temperature \( T_{\text{ad}} \) (changing with atmospheres) after the onset of ignition. The heating rate is a crucial quantity to understand pulverized coal combustion for single particles. In the following, different effects on the homogeneous ignition and volatile combustion are discussed with respect to the observations noticed in experiments and simulations.

6.1. Effect of particle size

With an increasing particle diameter \( d_p \), both \( t_{\text{ign}} \) and \( t_{\text{vol}} \) evidently increase, as shown in the experimental results of Fig. 14 and Fig. 15. In the aforementioned validation, simulations properly captured this behavior as one can observe in Fig. 17.
is then of interest to understand the effects of particle size on single particle combustion. Considering the energy balance in Eq. 3, the heating rate decreases with increasing \(d_p\) if other parameters remain unchanged. To further explain this phenomenon, the particle temperature histories of differently sized particles are numerically evaluated for cases C3 to C6 under the condition OXY30. Figure 18(a) shows an evidently lower slope in the rise of \(T_p\) and delayed ignition (dashed lines) for larger particle diameters. Additionally, the local gas temperature \(T_{g,local}\) is calculated by averaging \(T_g\) within a volume of 1 mm\(^3\) centered around the particle. In Fig. 18(b), an apparent decrease of \(T_{g,local}\) at the early heating stage is noticed. The thermal energy stored within the gas phase is responsible for the heat supply for the evaporation of moisture, increasing the particle temperature and volatile release. As heat transfer from the gas phase to the particle occurs, the gas phase temperature close to the particle is lowered, therefore convective heat transfer, which scales linearly with the temperature difference, is decreased. Due to the higher thermal inertia of larger particles, more energy is required from the gas phase for the particle heat-up, which leads to a more significant gas temperature drop as one can see in Fig. 18(b). Consequently, a lower local gas temperature leads to a further decrease of the heating rate, as indicated in Eq. 3. Moreover, the subsequent devolatilization process is essentially impacted by the initial particle heating. The volatile release rate can be depicted by the temporal evolution of the particle mass loss ratio, \(m_p/m_{p,0}\), where \(m_p\) and \(m_{p,0}\) denote the instantaneous and initial particle mass respectively. Figure 18(c) indicates a temporally retarded volatile release with increasing particle size. Similar with \(t_{ign}\), the increase of \(t_{vol}\) can be explained by (1) the longer devolatilization due to the lower heating rate as discussed above and (2) the higher volatile mass leading to an increase of the entire duration for the release of hydrocarbons and their consumption through combustion.

6.2. Effect of oxygen concentration

In experiments, it has been observed under both AIR and OXY conditions, that the ignition delay time decreases with increasing oxygen concentration as seen in Fig. 14. In other experimental studies, similar trends were observed by means of time-averaged CH\(^*\) imaging measurements [13] and single-shot sooting flame imaging measurements [14]. An explanation can be found in the molecular diffusion rate, which is proportionally scaled to the concentration gradient. Thus, a higher oxygen partial pressure promotes the formation of a flammable gas mixture inducing auto-ignition. Besides, an increased \(O_2\) content leads to a higher oxygen atom concentration close to the particle and hence further increased reactivity of fuel-oxidizer mixtures. Ignition preferentially occurs under lean conditions [20, 25, 44] for which
higher oxygen concentrations in the atmosphere are advantageous. An analysis of the mixture fraction field from simulations is expected to provide more insights in future investigations.

Further, increasing oxygen concentrations result in a shorter duration of volatile combustion. The same effect has been investigated using particle temperature histories measured by optical pyrometry [8, 9] or by classifying soot images to derive volatile combustion durations [14]. An accelerated devolatilization process can be argued for higher oxygen concentration with several evidences as follows. First, fuel oxidation in volatile diffusion flames benefits from faster oxygen diffusion due to a higher partial pressure. Moreover, sooting flame temperatures increase in O$_2$-enriched environments [8, 11, 12, 14, 15]. A higher gas temperature (1) increases the diffusivity of fuel and oxidizer and (2) increases the heating rate which expedites volatile release as well (e.g. see Eq.3). Furthermore, in the presence of more O$_2$, the flame size reduces [9, 12] and the reaction zone is located closer to the particle surface [18], which increases the heating rate through thermal conduction.
6.3. Effect of slip velocity

As the slip velocity, defined as the relative velocity between the particle and the gas phase, is difficult to examine experimentally, its effect is hereafter explored by means of numerical simulation. The cases C_7 and C_8 using artificially increased slip velocities are compared with the cases C_3 and C_4 with respect to \( t_{\text{ign}} \), \( T_p \), and \( T_{g,\text{local}} \). Figure 19(a) shows that increased slip velocities decrease the ignition delay time. Two effects might explain this behavior:

1. accelerated convective heating due to an increased Nusselt number \( Nu \), and,
2. accelerated convective heating due to a higher local gas phase temperature as the particle moves through the domain and draws energy from the gas phase.

Due to the dependency of forced convection on velocity, one can assume that an increased relative velocity between particle and gas phase results in a more efficient heat transfer to the particle. Indeed, convective heat transfer from the Eulerian to the Lagrangian phase scales linearly with the Nusselt number. In the present numerical setup, the Nusselt number is computed as

\[
Nu = 2 + 0.552 \sqrt{Re_p Pr^3},
\]

where \( Pr = 0.7 \) following calculations by Ranz and Marshall [46]. However, from Eq. 4 it can be seen, that for a realistic range of the particle Reynolds number \( Re_p \), the effect of the slip velocity on the Nusselt number is small. Therefore, the major contribution to increased heating rates for high slip velocities leading up to combustion is the higher temperature of the gas field surrounding the particle. In the pre-ignition stage, water evaporation and particle heating causes a temperature decrease in the vicinity of the particle. With increasing slip velocities, heat transfer from the gas phase to the particle is more efficient due to the ongoing exchange of the adjacent gas phase. As heat is drawn from the surrounding gas phase, a higher slip causes a faster replacement of the cooled-down gas with hot gas upstream of the particle, therefore establishing an overall higher temperature difference which induces a faster heating of the particle. Figure 19 illustrates this effect for both slip-induced increased particle temperatures \( T_p \) in Fig. 19(b) and the simultaneous cool-down of the adjacent gas phase for high slip velocities in Fig. 19(c). Subsequently, volatile release is expedited because of higher slip velocities and therefore higher heating rates which eventually causes earlier ignition as shown in Fig. 19(a). The positive effect of slip velocity on particle ignition was observed in the previous work with a similar configuration [47]. It was concluded that the ignition delay time was longer when assuming zero slip velocity.
After reaching the maximum volatile release rate, e.g. at 15 ms for particles with a diameter of 120 µm, the majority of volatiles have already been released and the particle has lost approximately 35% of its initial mass as can be seen in Fig. 12(c). At this stage, the heat release of the surrounding volatile diffusion flame has increased the local gas temperature above its initial value. Therefore, high slip now promotes convective heat loss, which is visible in the stronger decrease of the local gas phase temperature \( T_{g,\text{local}} \) after reaching its peak in Fig. 19(c). Consequently, the situation is inverted as the heating rate of particles with higher slip velocity is now lower. In addition, a higher \( U_s \) promotes gas species transport by enhancing convection, which results in faster replacement of the remaining combustible gas species fuels around the particle with the non-reactive surrounding gas. This results, amongst other effects, in an earlier decrease in the CH mass fraction and as a result in a decrease of the volatile combustion duration as one can observe in Fig. 19(a).

6.4. Effect of CO\(_2\)

In the literature, different experiments [11, 13, 14] on the single-particle scale have been conducted indicating that the replacement of N\(_2\) with CO\(_2\) delays particle ignition. This is usually explained based on the molar heat capacity (or so-called heat sink, \( \rho c_p \)) [4, 5, 13, 14], which is sufficiently larger for CO\(_2\) compared to N\(_2\) (e.g. a ratio CO\(_2\)/N\(_2\) = 1.7 at 1400K). In this study, we observe that the differences of the ignition delay time between N\(_2\) and CO\(_2\) atmospheres are minor for small particles, as can be seen in Fig. 14. For large particles with \( d_p = 205 \mu\text{m} \), \( t_{\text{ign}} \) is equivalent or even slightly decreased in the presence of CO\(_2\). A delay in the ignition time with N\(_2\) replaced by CO\(_2\) is not observed in the present work. Based on this experimental observation, theoretical and numerical foundations are discussed as follows.

For auto-ignition following the classical thermal explosion problem, the ignition delay time \( \tau_1 \) of a reactant gas mixture can be expressed as [48]:

\[
\tau_1 = \frac{c_v T_0^2 / T_a}{q_c Y_{F,0} A \exp(-T_a/T_0)},
\]

where \( c_v \) denotes the specific heat capacity of the mixture, \( q_c \) is the reaction heat release rate, \( T_0 \) is the initial temperature, \( T_a \) is the activation temperature, \( Y_{F,0} \) is the fuel mass fraction, and \( A \) is the pre-exponential factor for reaction kinetics. It is commonly argued that the higher \( c_v \) of CO\(_2\) slows down the temperature rise and hence chemical reaction leading to a larger ignition delay time. However, it is important to note, that the impact of \( c_v \) cannot be solely considered when introducing CO\(_2\), because the ignition delay time is a function of multiple parameters.

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Figure 19: Effect of slip velocity. (a) Ignition delay time and volatile combustion duration, (b) particle temperature and (c) local gas temperature evolution for two different particle sizes in OXY30 condition. The cases $C_3$, $C_4$, $C_7$, and $C_8$ are included, and the corresponding $t_{\text{ign}}$ are indicated by dashed lines in (b) and (c).

It is difficult to systematically explore the changes that CO$_2$ introduces by only exchanging the atmosphere, even under fully-defined conditions in the present experiments, since e.g. the flow rates for all CO$_2$ atmospheres are reduced to obtain gas temperature profiles similar to N$_2$ atmospheres. As a consequence, the particle slip velocity is lower in OXY conditions, as shown in Fig. 12 which is another parameter crucially influencing the ignition and combustion behavior of solid fuel particles. With the aid of simulations, it has been discussed above that a higher slip velocity increases heat convection and hence shortens ignition delay if other boundary conditions remain unchanged. In other words, assuming the same slip velocity as in AIR conditions, $t_{\text{ign}}$ in OXY conditions (see Fig. 14(b)) would further decrease lead-
ing to more significant differences between N\textsubscript{2} and CO\textsubscript{2} atmospheres. In summary it can be said, that an experimental study of the influence of CO\textsubscript{2} always introduces changes in either thermodynamics (i.e. temperatures) or aerodynamics (i.e. slip velocities) when trying to maintain equal conditions for both AIR and OXY atmospheres. To overcome this inherent problem, this study employs a combined experimental-numerical approach.

As illustrated in Fig. 20, a reasonable comparison can only be drawn with identical slip velocities and initial gas temperatures. Here, the numerical results of cases C\textsubscript{1}, C\textsubscript{2}, C\textsubscript{7}, and C\textsubscript{8} are compared. During the particle heating, heat transfer (convective, conductive and radiative) from the gas phase to the solid phase leads to a drop in the gas temperature in the vicinity of particles, as shown in Fig. 20(a). Considering the same amount of heat extracted, the gas temperature decreases less in CO\textsubscript{2} than in N\textsubscript{2} due to a higher \(c_v\). For the same particle diameter, it can be seen that the local gas temperature \(T_{g,\text{local}}\) is higher in the presence of CO\textsubscript{2} than within N\textsubscript{2} atmospheres, especially during the particle heating. This discrepancy is more evident for a larger particle diameter of \(d_p = 205\mu m\) since more overall energy is required for heating. A higher local gas temperature increases the particle heating rate before ignition, as indicated in Eq. 3. In addition, the thermal conductivity of CO\textsubscript{2} is about 1.2 times higher than that of N\textsubscript{2} at 1400 K [4]. Hence, an earlier volatile release and formation of an ignitable fuel mass fraction \((Y_{F,0})\) can be expected. This is verified by computing the maximum fuel mass fractions \(Y_{C_2H_2}+Y_{CH_4}\) in the vicinity of the particle as a representative \(Y_{F,0}\) for volatile combustion. In Fig. 20(b), an early increase of fuel mass fraction is noticeable in the CO\textsubscript{2} atmosphere, which also implies an early formed flammable gas mixture. Equation 5 indicates that the gas temperature \(T_0\) (here equivalent to \(T_{g,\text{local}}\)) and \(Y_{F,0}\) have opposite effects compared with \(c_v\) or \(\rho c_v\) on the ignition delay time. For example, Khatami et al. [15] explained in DTF measurements that the retarded ignition in CO\textsubscript{2} is mainly attributed to the lower ambient temperature compared to that in N\textsubscript{2} in their facility. Since differences of \(T_{g,\text{local}}\) at ignition are minor in this study, as indicated by Fig. 20(a), the fuel mass fraction is considered as a dominant effects probably inducing early ignition. In summary, based on the present study which has been performed at an initial gas temperature of around 1800 K, combining experiments and simulations, the replacement of N\textsubscript{2} with CO\textsubscript{2} does not necessarily result in a delayed ignition with the same initial global gas temperature. In general, CO\textsubscript{2} introduces competing effects on the ignition delay time via the changes of the local gas temperature and the heat capacity. It is important to emphasize that auto-ignition is a local phenomenon, which requires an examination of the possible influence factors (i.e. in Eq. 5) in a relevant domain. Although it is strongly impacted by global boundary conditions, the local
heating conditions and gas mixture fraction are dominating factors and need to be considered carefully.

Figure 20: Effect of CO$_2$. (a) Local gas temperature $T_{g,\text{local}}$ histories and (b) the temporal change of the maximum fuel mass fraction (represented by $Y_{C_2H_2} + Y_{CH_4}$) of two different particle sizes during the ignition under AIR30 and OXY30 conditions. The cases C$_1$, C$_2$, C$_7$, and C$_8$ are compared, and the corresponding $t_{\text{ign}}$ are indicated by dashed lines.

Previous experimental [14, 15] and numerical [22, 49] investigations have reported convincing evidences of a longer volatile combustion duration in CO$_2$ atmospheres. In accordance with literature, in Fig. 15 and Fig. 17, the increase of the volatile combustion duration is observed in both experimental and numerical data, which appears more clearly for larger particles. Considering the volatile flame as a typical diffusion flame, there are several explanations. Above all, gas diffusion is retarded by replacing N$_2$ with CO$_2$. The diffusivity of O$_2$ into CO$_2$ is 0.75 times that into N$_2$ at 1400 K and light hydrocarbons like CH$_4$ also show a lower diffusivity in CO$_2$. In addition, the peak flame temperatures are lower in CO$_2$ due to the high heat capacity. This effect has been reported in [14, 15] and is also evident according to our results in Fig. 20. Moreover, the O$_2$ diffusivity is a function of temperature which decreases further in CO$_2$ atmospheres. Although the effect might be minor, the entire volatile yield is reported to be higher in CO$_2$ atmospheres. In conclusion, a retarded gas diffusion and an overall slowed fuel consumption rate can be expected when replacing N$_2$ with CO$_2$. 

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7. Conclusions

The present work reports a detailed investigation of single particle combustion of a Colombian hvb coal under conventional and oxy-fuel atmospheres using a combined experimental-numerical approach. The laminar flow reactor was comprehensively characterized by a series of experiments e.g. with PIV and quantitative OH-LIF, providing fully defined boundary conditions of e.g. inlet gas compositions, outlet gas temperature and velocity profiles, particle heating points (i.e. the position of the flame front), and particle size and shape. Two particle samples A (mean \( d_p = 120 \mu m \)) and B (mean \( d_p = 205 \mu m \)) were investigated in four \( \text{N}_2 \) atmospheres (containing 10vol% - 40vol% \( \text{O}_2 \)) and three \( \text{CO}_2 \) (containing 20vol% - 40vol% \( \text{O}_2 \)) atmospheres. Multi-parameter optical measurements enabled the simultaneous detection of the ignition delay time \( t_{\text{ign}} \) and the volatile combustion duration \( t_{\text{vol}} \) for each individual coal particle. Time-resolved OH-LIF revealed a high sensitivity and accuracy for ignition detection, while broad-band luminescence imaging over-predicted homogeneous ignition with a delay of \( 3 \sim 5 \text{ms} \) on average. Numerical simulations performed in an Eulerian-Lagrangian framework were validated against experimental results and further provided insights into multi-phase heat transfer and volatile release by computing essential quantities of e.g. \( T_p, T_{\text{g,local}}, m_p/m_{p,0} \) and \( Y_F \). By analyzing experimental and numerical observations and including theoretical evidences, the effects of particle size \( d_p \), \( \text{O}_2 \) enrichment, slip velocity \( U_s \), and \( \text{CO}_2 \) addition were discussed with regard to \( t_{\text{ign}} \) and \( t_{\text{vol}} \). Several conclusions are summarized as follows:

1. Both \( t_{\text{ign}} \) and \( t_{\text{vol}} \) significantly increase with the particle diameter \( d_p \). The retarded ignition is attributed to a decreased particle heating rate, which is associated with a larger \( d_p \) and lower \( T_{\text{g,local}} \) induced by heat transfer between gas phase and particle. The increased volatile combustion duration mainly results from a higher volatile content stored in larger particles which eventually extends the devolatilization period, whereas the different particle heating rates before the onset of ignition might have minor influences on \( t_{\text{vol}} \).

2. Both \( t_{\text{ign}} \) and \( t_{\text{vol}} \) decrease with an increasing \( \text{O}_2 \) concentration in all investigated atmospheres, with a more pronounced effect for larger particles. The higher partial pressure of \( \text{O}_2 \) accelerates the formation of a flammable gas mixture (i.e. shortens \( t_{\text{ign}} \)) and raises the fuel consumption rate (i.e. shortens \( t_{\text{vol}} \)) by faster molecular diffusion. For \( t_{\text{vol}} \), the \( \text{O}_2 \)-dependent flame temperature and flame size are considered to be of secondary importance.

3. Higher slip velocity accelerates the heat convection between the gas phase and the particle due to a faster replacement of the cooled down gas around the par-
ticle with hot gas upstream of the particle, which eventually reduces $t_{\text{ign}}$. After releasing the majority of volatiles, heat loss and molecular transport by convection close to the particle are enhanced by an increased $U_s$, and accordingly, $t_{\text{vol}}$ shows a slight tendency to decrease due to a higher fuel consumption rate.

4) With an identical $U_s$ and global $T_g$, CO$_2$ mitigates the decrease in $T_{g,\text{local}}$ caused by evaporation and particle heating in the pre-ignition stage due to its higher heat capacity compared with N$_2$. This subsequently results in an early release of light hydrocarbons and flammable gas mixtures promoting auto-ignition. This effect is more significant for larger particles, which is seen in both simulations and experiments. On the other hand, volatile combustion is retarded by replacing N$_2$ with CO$_2$, which is explained by the lower diffusivity of both O$_2$ and light hydrocarbons within CO$_2$.

Acknowledgements

The authors kindly acknowledge financial support through Deutsche Forschungsgemeinschaft (DFG) – Projektnummer 215035359 – TRR 129 for its support through CRC/Transregio 129 “Oxy-flame: development of methods and models to describe solid fuel reactions within an oxy-fuel atmosphere.” Gefördert durch die Deutsche Forschungsgemeinschaft (DFG) - Projektnummer 215035359 - TRR 129.

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