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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 O₂ 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 O₂ 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 N₂ to CO₂ 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 CO_2 , 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 CO_2 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

1 Graphical abstract

2 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 N_2 replacement by CO_2
- The particle size and gas composition dominate the ignition delay time via affecting the particle heating rate

- 12 • The N₂ replacement with CO₂ will not delay the ignition if the global gas
13 temperature is constant

Nomenclature

Greek letters

β	aspect ratio (-)
ρ	gas density (kg/m ³)
ρ_p	particle density (kg/m ³)
ϕ_k	distribution coefficient (-)
Φ	mixture fraction (-)
ν	kinematic viscosity (m ² /s)
ν_{ji}	central wavelength (nm)
ϵ	area estimation error (-)
ϵ_p	particle emissivity (-)
ϵ_g	gas emissivity (-)
τ_i	ignition delay time of gas mixtures (ms)
σ	Stefan-Boltzmann constant (-)
Δt	time difference (ms)

Symbols

a	ellipse major axis (mm)
A	integral absorption (-)
A	pre-exponential factor for reaction kinetics (-)
A_e	area of a ellipse (mm ²)
A_p	particle area (mm ²)
A_{LU}	area of luminescence signals (mm ²)
b	ellipse minor axis (mm)
$B_{j,i}$	Einstein B coefficient (-)
c_p	heat capacity (constant pressure) (J/(kg·K))
c_v	heat capacity (constant volume) (J/(kg·K))
d_{90}	diameter of 90th percentile (μm)
d_p	circle-equivalent particle diameter (μm)
d_x	length of cubic cells (μm)
D_i	degeneracy factor (-)
f	focal length (mm)
$f_{B,i}(T)$	temperature dependent Boltzmann fraction (-)
Fo	Fourier number (-)

g_0	spectral line overlap integral (nm)
h	convection coefficient of heat transfer (-)
I_{LIF}^*	absorption-corrected LIF signals (a.u.)
L_d	distribution length (m)
m_p	instantaneous particle mass (kg)
$m_{p,0}$	initial particle mass (kg)
Nu	Nusselt number (-)
Pr	Prandtl number (-)
q_c	the reaction heat release rate (J/s)
Re_p	particle Reynolds number (-)
t_{ign}	particle ignition delay time (ms)
$t_{\text{i,LU}}$	first appearance of luminescence signal (ms)
t_{vol}	volatile combustion duration (ms)
$t_{\text{vol,end}}$	end time of volatile combustion (ms)
t	residence time (ms)
T	optical transmission (-)
T_a	activation temperature (K)
T_{ad}	adiabatic flame temperature (K)
T_g	gas temperature (K)
$T_{g,\text{local}}$	local gas temperature (K)
T_0	initial global gas temperature (K)
U_g	axial gas velocity (m/s)
U_p	axial particle velocity (m/s)
U_s	axial slip velocity (m/s)
Y_{CH}	CH mass fraction (-)
$Y_{\text{C}_2\text{H}_2}$	C ₂ H ₂ mass fraction (-)
Y_{CH_4}	CH ₄ mass fraction (-)
Y_{F}	fuel mass fraction (-)
$Y_{\text{F},0}$	initial fuel mass fraction (-)
Y_{OH}	OH mass fraction (-)

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

FF	flat flame
FOV	field of view
FWHM	full width half maximum
hvb	high volatile bituminous
LFR	laminar flow reactor
LIF	laser-induced fluorescence
LU	luminescence
PDF	probability density function
PIV	particle image velocimetry
PTV	particle tracking velocimetry
ROI	region of interest
SAS	signal and structure
SNR	signal-to-noise ratio
SPC	single particle combustion
UV	ultraviolet

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 (CO_2) emissions, oxy-fuel combustion is a promising technology for coal-fired power generation. This approach uses a mixture of pure oxygen (O_2) as an oxidizer combined with recirculated flue gas which results in a replacement of nitrogen (N_2) in air to CO_2 . Consequently, the flue gas mainly contains CO_2 and water (H_2O) which facilitates CO_2 capture and storage. However, the combustion processes are impacted significantly by introducing CO_2 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.

1.1. 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 (hvb) coal in an entrained-flow reactor (EFR) with ambient gas temperatures T_g of ~ 1250 K [13]. Time-averaged CH^* signals recorded with an intensified CCD camera were used to derive the ignition delay time t_{ign} and the volatile combustion duration t_{vol} . It was concluded that O_2 enrichment significantly reduces t_{ign} . Further, the presence of CO_2 retards t_{ign} of single coal particles, whereas the volatile combustion duration t_{vol} is affected marginally. They reasoned that the increase in the volumetric heat capacity (the heat sink, ρc_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_{ign} and t_{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 N_2 and CO_2 atmospheres. Moreover, both t_{ign} and t_{vol} seemed to be greater when N_2 is replaced by CO_2 . 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 O_2 mole fraction, t_{ign} decreases significantly in a CO_2 -enriched environment, while remaining almost unaffected in an N_2 -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 ~ 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 O_2 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

108 aspects including the sensitivity of ignition delay time to temperature, gas compo-
 109 sition, particle size, and particle number density. The results have been validated
 110 against a series of experiments in a laminar entrained flow reactor [24]. A double
 111 peak profile in the time evolution of the volatile release rate has been observed,
 112 which strongly affects the ignition delay time. It has been shown that ignition can
 113 occur on the onset of the first or the second peak of the devolatilization rate in dif-
 114 ferent initial gas temperatures and particle sizes. It has also been shown that particle
 115 Reynolds number and slip velocity between gas and particles have an impact on ig-
 116 nition delay time and the volatile flame which cannot be neglected. The formation
 117 of wake flames due to high slip velocities becomes increasingly relevant for flame
 118 interactions with multiple interacting particles [25] and turbulent coal combustion
 119 [26]. Switching from conventional to oxy-fuel combustion can affect the combustion
 120 behavior due to the different thermal and chemical properties of CO_2 compared with
 121 N_2 . For instance, several numerical [27, 28, 29] studies indicate, that replacing N_2 by
 122 CO_2 increases the ignition delay time and the duration of devolatilization.

123 *1.3. Objective of the present work*

124 The present work's aim is the investigation of relevant physico-chemical effects on
 125 pulverized coal combustion on a single particle scale. For this purpose, the ignition
 126 and volatile combustion of single bituminous coal particles are examined employ-
 127 ing both experimental and numerical methods under well-defined laminar flow con-
 128 ditions. Because consecutively performed measurements of different quantities can
 129 only be combined using statistics with the need for an evaluation of the repeatabil-
 130 ity of the investigated process, multi-parameter measurements are conducted in this
 131 study such that relevant quantities are measured simultaneously providing a com-
 132 prehensive data set with high spatial and temporal resolution. The volatile flame
 133 is visualized by using OH-LIF to detect the ignition and using luminescence imag-
 134 ing to determine the temporal end of volatile combustion. Additionally, high-speed
 135 backlight illumination addresses particle size, shape and velocity. With well defined
 136 boundary conditions, experimental results are analyzed showing the importance of
 137 particle size and ambient gas composition. Detailed numerical simulations are con-
 138 ducted and validated against experiments with respect to the ignition delay time and
 139 volatile combustion duration. Beyond that, simulations provide further insights into
 140 particle temperatures, local gas temperatures, volatile release rates, and fuel mass
 141 fractions. Combining experimental and numerical expertise, effects of particle size,
 142 oxygen concentration, slip velocity and the introduction of a CO_2 atmosphere are
 143 discussed to understand the ignition and volatile combustion in the present configu-
 144 ration.

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 80×80 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 $\text{CH}_4/\text{O}_2/\text{N}_2$ and three CO_2 atmospheres (denoted as OXY) with varying inlet mixtures of $\text{CH}_4/\text{O}_2/\text{CO}_2$ were investigated. Table 2 lists the oxygen concentration in the post-flame environment, flow rates, equivalence ratio Φ , and adiabatic flame temperatures T_{ab} for each condition. Based on the 1D calculation, the produced water vapor concentration was ~ 14 vol% and ~ 16 vol% in AIR and OXY conditions, respectively. Cai et al. [30] reported that particle ignition is not affected by a H_2O 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,

181 volatile combustion and char combustion. By modulating the wheel rotation speed
 182 of the seeding unit, individual particles were sparsely seeded to guarantee single par-
 183 ticle combustion. To give an impression of particle combustion in the LFR, luminous
 184 streaks of burning particles are shown in the color image of Fig. 1(b).

185 Colombian high-volatile bituminous coal particles with two different sievings were
 186 used in this study. The samples, denoted as A and B, were sieved to a range of
 187 90 to 125 μm and 160 to 200 μm , respectively. The proximate analysis of the coal
 188 composition was 3.5% moisture(an), 36.9% volatiles(wf), 54.4% Cfix(wf) and
 189 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.

O ₂ content		Φ (-)	Flat flame (ln/h)			Particle jet (mln/min)			T _{ad} (K)
			CH ₄	N ₂	O ₂	CH ₄	N ₂	O ₂	
AIR10	10 vol%	0.58	312	3203	1085	1.12	11.49	3.89	1847
AIR20	20 vol%	0.40	312	2744	1543	1.12	9.85	5.54	1839
AIR30	30 vol%	0.31	311	2286	2003	1.12	8.21	7.18	1829
AIR40	40 vol%	0.25	313	1822	2286	1.12	6.54	8.84	1829
			CH ₄	CO ₂	O ₂	CH ₄	CO ₂	O ₂	
OXY20	20 vol%	0.47	307	1874	1315	1.44	8.86	6.19	1835
OXY30	30 vol%	0.37	306	1553	1672	1.43	7.25	7.81	1881
OXY40	40 vol%	0.29	305	1310	2094	1.36	5.83	9.31	1825

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_2O_3 particles (MR-52, Martoxid, $d_{90} = 3 \sim 6 \mu\text{m}$) 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 $\sim 100 \mu\text{m}$. 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 $20 \mu\text{m}$.

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 16×16 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 in-
 tegral absorption was calibrated using spectroscopic data from LIFBASE [33] using
 Eq. 1,

$$\int [OH](x, y) dr = \frac{A(y)}{g_0 \frac{h\nu_{ji}}{c} B_{ji} D_i f_{B,i}(T)}, \quad (1)$$

where A denotes the measured integral absorption, g_0 the spectral line overlap in-
 tegral, ν_{ji} 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 ab-
 sorption 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.^{R2}

$$[OH](x, y) = \frac{I_{LIF}^*(x, y)}{\int I_{LIF}^*(x, y) dx} \int [OH](x, y) dx. \quad (2)$$

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 can-
 celed. This method yields a very high sensitivity and precision, as the equilibrium
 OH number density doubles approximately every 100 K under these conditions. How-
 ever, 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 $X^2\Pi \rightarrow A^2\Sigma$ ($\nu'' = 0 \rightarrow \nu' = 1$) 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

247 (WSU-30, HighFinesse) which was calibrated using a temperature stabilized single
 248 mode HeNe diode laser. The laser system was operated at 500 Hz repetition rate. OH
 249 fluorescence was detected using a high-speed CMOS camera (HSS6, LaVision) cou-
 250 pled with a two-stage image intensifier (HS-IRO, LaVision). A 100 mm lens (2178,
 251 Cerco, $f/2.8$) was employed for high UV-transmission. A narrow-band band-pass fil-
 252 ter ($T \geq 90\%$ @ 310 – 320 nm) was used to suppress ambient light as well as Rayleigh
 253 scattering. The filter transmits only the (1,1) vibrational band to minimize signal re-
 254 absorption [34]. For the absorption measurement, a fraction of the laser beam before
 255 and after the probe volume was extracted and guided to a reference cell filled with
 256 a diluted Rhodamin 6G ethanol solution. Beam steering effects were minimized by
 257 introducing a 4f-imaging lens in the post-burner beam path. Fluorescence was de-
 258 tected using a second high-speed camera (Fastcam SA-X2, Photron) equipped with
 259 a 180 mm lens (Sigma, $f/5.6$).

Figure 3: Gas temperature field characterization: experimental setup for the combined OH-PLIF/absorption spectroscopy measurements.

260 2.3. Multi-parameter measurements of single particle combustion

261 Simultaneous 10-kHz multi-parameter optical measurements were performed in-
 262 cluding planar OH-LIF imaging, diffuse-backlight illumination (DBI) and lumines-
 263 cence (LU) imaging. Figure 4 illustrates the experimental setup including the ar-
 264 rangement of the three diagnostic systems. For the reaction zone visualization using
 265 OH-LIF, the dye laser system introduced for the temperature measurements was
 266 employed at a repetition rate of 10 kHz. The wavelength was tuned to 283.01 nm
 267 to excite the $Q_1(6.5)$ line of the A-X(1-0) transition system of the OH radical. The
 268 effective pulse energy in the probe volume was approximately 0.3 mJ. The laser beam
 269 was formed into a sheet with a thickness of 0.8 mm (FWHM) at the burner center
 270 line. The aim of this relatively thick laser sheet was to reduce the probability of out-
 271 of-plane loss particles and to allow the signal detection in the early stage of volatile
 272 flames. The emission signals were collected by the aforementioned high-speed intensi-
 273 fied camera (HSS6 + HS-IRO, LaVision) equipped with a UV-achromatic camera lens

274 (Halle, $f = 150 \text{ mm}$, $f/2.5$). To suppress broadband chemiluminescence and thermal
 275 radiation from volatile flames, a band-pass filter ($T \geq 40\%$ @ $305\text{--}340 \text{ nm}$) was used
 276 and the intensifier gate was shortened to 100 ns . The field of view (FOV) for OH-LIF
 277 imaging was $19 \times 19 \text{ mm}^2$ with a projected pixel size of $25 \mu\text{m}$.

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

286 To track the temporal evolution of burning particles, another CMOS camera
 287 (Fastcam SA-X2, Photron) combined with a macro lens (Sigma, $f = 180 \text{ mm}$, $f/5.6$)
 288 was employed to image the luminous flame. A band-pass filter ($380\text{--}492 \text{ nm}$, Sem-
 289 rock) was used and the recorded intensity was dominated by the thermal radiation
 290 of soot, tar and particle surface [18]. The FOV of $65 \times 10 \text{ mm}$ (height $\times$ width) al-
 291 lowed a particle tracking throughout the entire volatile combustion stage. Due to the
 292 large field of view, the projected pixel resolution was restricted to $60 \mu\text{m}$ but was still
 293 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.

294 2.4. Multi-parameter data processing

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

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

303 As one of the key quantities, the ignition delay time t_{ign} is determined by tem-
 304 porally and spatially tracking the OH-LIF signal. Instantaneous laser profiles are
 305 obtained from the background OH-LIF intensity associated with the post-flame re-
 306 gion, where a homogeneous distribution of OH-radicals from water dissociation is
 307 assumed. The OH-LIF images are shot-for-shot corrected in terms of the pulse en-
 308 ergy fluctuation and the sheet inhomogeneity. The signal-to-noise ratio (SNR) is
 309 estimated to be approximately 12 and the maximum signal-to-background ratio is
 310 around 10. The signals are normalized based on the background intensity and further
 311 denoised by using a 5×5 Gaussian filter. The particle centroids are extracted from si-
 312 multaneous DBI recordings and used as position references for particle tracking. Only
 313 the particles with a sufficient distance of ≥ 4 mm to the nearest particle were included
 314 in the data analysis which guaranteed single-particle combustion mode. As shown in
 315 Fig. 5 (a,c,d), a 2×2 mm² region of interest (ROI, dashed line) is defined centered
 316 around the particle centroid (red cross) in the pre-processed OH-LIF images. By
 317 evaluating the background intensity, an intensity threshold of 1.2 is applied for bi-
 318 narization in Fig. 5 (b,d,e). Within the ROIs, a signal and structure (SAS) analysis
 319 was performed. The ignition time is determined to when (1) the mean OH-LIF in-
 320 tensity increases over the threshold and (2) the connected area of the largest binary
 321 structure exceeds 1×1 mm². The latter criterion is defined based on an estimation
 322 of the spatial resolution restricted to the laser sheet thickness. Figure 5 illustratively
 323 shows three instants at $t_{\text{ign}} - 0.5$ ms, t_{ign} and $t_{\text{ign}} + 0.5$ ms for a single-particle ignition
 324 event. The heating start time is set to $t = 0$ and is defined at the instant at which
 325 a particle crosses the flame front of the premixed CH₄ flame.

326 The volatile flame duration t_{vol} is determined by combining t_{ign} from OH-LIF
 327 measurements and the end time of volatile combustion $t_{\text{vol,end}}$ from luminescence
 328 measurements. The particle positions extracted from DBI images are used to initial-
 329 ize the temporal tracking of LU signals. The area of LU signals A_{LU} is evaluated by
 330 defining an intensity threshold which is placed marginally (i.e. $\sim 10\%$) above the
 331 background level. Figure 6(a) shows the temporal variation of the LU signal area
 332 of a single particle flame. Four individual instants are highlighted and visualized in
 333 Fig. 6(b-e). The color scale has been adapted to match the low intensity level. A
 334 few milliseconds after the onset of ignition, the size of the gas flame increases and
 335 then decreases to a local minimum. This implies a transition from the formation of
 336 a diffusion flame to the completed fuel consumption via gas-phase oxidation [18]. In

Figure 5: An example of ignition detection using temporal OH-LIF signals at (a, b) 0.5 ms before ignition, (c, d) ignition and (e, f) 0.5 ms after ignition. Left column: pre-processed OH-LIF images superposed with particle centroids (red crosses) from simultaneous DBI images and a $2 \times 2 \text{ mm}^2$ region of interest (dashed lines). Right column: corresponding binarized OH-LIF images.

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 $t_{\text{vol, end}}$. In a few cases, the luminosity intensity disappears at the end of volatile combustion and appears after char combustion begins. The $t_{\text{vol, end}}$ 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 $t_{\text{vol}} = t_{\text{vol, end}} - t_{\text{ign}}$.

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]. The

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 P_1 and P_2 have a similar shape (β), whereas particle P_2 and P_3 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

Figure 7: DBI images of there representative particles and definitions of relevant parameters. d_p : circle-equivalent diameter, β : aspect ratio of the fitted ellipse, A_p : particle area, A_e : area of the fitted ellipse, ϵ : error of the particle area approximated by the ellipse fitting.

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_2O , and other gases. Other gases are assumed to be C_2H_2 , similar to the assumption by Jimenez and Gonzalo-Tirado [29]. The rate of tar release, which is

also computed by the CPD model, is assumed to be only for C_2H_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 C_6H_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 C_2H_2 in a purely gas phase setting is much lower than that of C_6H_6 , the marginal difference in t_{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 ϕ_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 $125d_x^3$) 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 β (b) and the error of the DLSF approximation ϵ (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 $y = 2$ 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 μm and 205 μm for A and B respectively. For β and ϵ , no significant discrepancy between differently sieved particles is noticeable. The highest probability of β 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 $y = 2.5$ 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 $y = 7.5$ mm is observed for all conditions. Downstream of this, T_g decreases linearly with increasing y with a slope of ~ 2 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 β and the error of the DLSF approximation ϵ .

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

CH₄ 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 O₂ 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.

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

489 along y . As Figure 11 illustrates, the smaller particles of sample A show a higher
 490 initial velocity after crossing the flame front due to their lower mass inertia compared
 491 to the larger particles of sample B. Afterwards, the velocity slope of both particle
 492 sizes is similar, however, smaller particles have a higher acceleration considering the
 493 shorter residence time compared with larger particles. This points to the fact that
 494 the particle velocity history is essentially dominated by the steep gas velocity gra-
 495 dient within the flat flame, while the aerodynamic drag downstream is of secondary
 496 importance.

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 $d_p = 205 \mu\text{m}$).

497 With knowledge of the mean gas and particle velocities, the mean slip velocity U_s
 498 can be calculated and is depicted in Fig. 12 for all atmospheres and both size sam-
 499 ples. U_s first increases over the CH_4 flame and then decreases with the stabilization
 500 of the gas velocity after $y = 3 \text{ mm}$. The particle Reynolds number can be estimated
 501 using the slip velocity and the particle diameter to $Re_p = U_s d_p / \nu$, where ν denotes
 502 the kinematic viscosity of the gas mixture. Based on the gas composition and the
 503 adiabatic flame temperature, ν is estimated to be $\sim 3.3 \times 10^{-4} \text{ mm}^2/\text{s}$ for AIR condi-
 504 tions and $\sim 2.7 \times 10^{-4} \text{ mm}^2/\text{s}$ for OXY conditions. Using the maximum slip velocity
 505 and mean particle diameter, the particle Reynolds number of small particles is esti-
 506 mated to be 0.29 in AIR and 0.17 in OXY conditions; the particle Reynolds number
 507 of large particles is 0.73 in AIR and 0.60 in OXY conditions.

508 4.4. Comparison of methods for ignition detection

509 Experimental results of ignition delay times always depend on the uncertainties
 510 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 $d_p = 120 \mu\text{m}$) and B (mean $d_p = 205 \mu\text{m}$).

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 $\Delta t = t_{i,LU} - t_{ign}$. Figure 13 shows PDFs of Δt 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 Δt 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 Δt 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

555 in Section 6.

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

556 4.6. Volatile flame duration

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

572 5. Numerical results

573 For the appropriate interpretation of experimental observations, detailed numer-
 574 ical 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 $d_p = 120 \mu\text{m}$) and B (mean $d_p = 205 \mu\text{m}$).

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 $y \approx 2 \text{ mm}$. 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.

Case	C_1	C_2	C_3	C_4	C_5	C_6	C_7	C_8
Atmosphere	AIR30		OXY30					
d_p [μm]	120	205	120	205	80	160	120	205
U_s [m/s]	0.81	1.17	0.39	0.79	0.2	0.6	0.81	1.17
T_g [K]	1829							

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

591 been quantified by taking the time at which 5% of the first peak in OH mass fraction,
 592 Y_{OH} , has been reached. This definition aims to reproduce the ignition detection of
 593 the experimental data, in which the first obvious OH signals above the background is
 594 detected. An example of the temporal change of Y_{OH} is shown in Fig. 16. This method
 595 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.

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

604 For larger particles, an over-prediction of the ignition delay time in the model
 605 is observed. Explanations for this deviation can be found in both measurements
 606 and simulations. In simulations, the over-prediction can be explained by Eulerian-
 607 Lagrangian modeling and the point particle approximation [44]. In the point particle
 608 approximation model, coal particles are assumed as points which represent homo-
 609 geneous spheres without any temperature gradient inside the particle which release
 610 volatile matter homogeneously in all directions. Defined by the CPD model, the de-
 611 volatilization occurs as the bonds in the internal structure of the coal particle break
 612 when exposed to heat at a certain particle temperature T_p which is essentially in-
 613 fluenced by the initial particle mass and the heating rate. At a high heating rate
 614 in the order of 10^5 K/s in the present work, the devolatilization has been experi-
 615 mentally observed as a surface phenomenon [45]. The Fourier number Fo of ~ 0.08
 616 and ~ 0.02 are estimated for smaller and larger particles, respectively, [20] which im-
 617 plies the existence of evident temperature gradients in the interior of particles. This

Figure 17: Comparison between numerical and experimental results with respect to t_{ign} and t_{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_{vol} is validated against experimental data in Fig. 17(b). t_{vol} is defined in the simulation by the time difference between

the ignition delay time and the time at which Y_{CH} decreases to 25% of its maximum value, shown in Fig. 16. This definition resembles the 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])

$$\frac{dT_p}{dt} = \frac{6}{d_p \rho_p c_p} (h(T_g - T_p) - \sigma(\varepsilon_g T_g^4 - \varepsilon_p T_p^4)), \quad (3)$$

where T_p , ρ_p , c_p and ε_p denote the temperature, density, heat capacity and emissivity of particles in order; h is the convection coefficient of heat transfer; T_g and ε_g are the temperature and emissivity of gases, respectively, and σ 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_{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_{ign} and t_{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. It

670 is then of interest to understand the effects of particle size on single particle com-
 671 bustion. Considering the energy balance in Eq. 3, the heating rate decreases with
 672 increasing d_p if other parameters remain unchanged. To further explain this phe-
 673 nomenon, the particle temperature histories of differently sized particles are numeri-
 674 cally evaluated for cases C₃ to C₆ under the condition OXY30. Figure 18(a) shows an
 675 evidently lower slope in the rise of T_p and delayed ignition (dashed lines) for larger
 676 particle diameters. Additionally, the local gas temperature $T_{g,local}$ is calculated by
 677 averaging T_g within a volume of 1 mm³ centered around the particle. In Fig. 18(b),
 678 an apparent decrease of $T_{g,local}$ at the early heating stage is noticed. The thermal
 679 energy stored within the gas phase is responsible for the heat supply for the evapo-
 680 ration of moisture, increasing the particle temperature and volatile release. As heat
 681 transfer from the gas phase to the particle occurs, the gas phase temperature close to
 682 the particle is lowered, therefore convective heat transfer, which scales linearly with
 683 the temperature difference, is decreased. Due to the higher thermal inertia of larger
 684 particles, more energy is required from the gas phase for the particle heat-up, which
 685 leads to a more significant gas temperature drop as one can see in Fig. 18(b). Conse-
 686 quently, a lower local gas temperature leads to a further decrease of the heating rate,
 687 as indicated in Eq. 3. Moreover, the subsequent devolatilization process is essentially
 688 impacted by the initial particle heating. The volatile release rate can be depicted by
 689 the temporal evolution of the particle mass loss ratio, $m_p/m_{p,0}$, where m_p and $m_{p,0}$
 690 denote the instantaneous and initial particle mass respectively. Figure 18(c) indicates
 691 a temporally retarded volatile release with increasing particle size. Similar with t_{ign} ,
 692 the increase of t_{vol} can be explained by (1) the longer devolatilization due to the
 693 lower heating rate as discussed above and (2) the higher volatile mass leading to an
 694 increase of the entire duration for the release of hydrocarbons and their consumption
 695 through combustion.

696 6.2. *Effect of oxygen concentration*

697 In experiments, it has been observed under both AIR and OXY conditions, that
 698 the ignition delay time decreases with increasing oxygen concentration as seen in
 699 Fig. 14. In other experimental studies, similar trends were observed by means of
 700 time-averaged CH* imaging measurements [13] and single-shot sooting flame imag-
 701 ing measurements [14]. An explanation can be found in the molecular diffusion rate,
 702 which is proportionally scaled to the concentration gradient. Thus, a higher oxy-
 703 gen partial pressure promotes the formation of a flammable gas mixture inducing
 704 auto-ignition. Besides, an increased O₂ content leads to a higher oxygen atom con-
 705 centration close to the particle and hence further increased reactivity of fuel-oxidizer
 706 mixtures. Ignition preferentially occurs under lean conditions [20, 25, 44] for which

Figure 18: Effect of particle size. (a) Particle temperatures T_p , (b) local gas temperatures $T_{g,local}$ and (c) particle mass loss $m_p/m_{p,0}$ histories for different particle sizes d_p of $80 \mu\text{m}$ (C₅), $120 \mu\text{m}$ (C₃), $160 \mu\text{m}$ (C₆), and $205 \mu\text{m}$ (C₄) in OXY30. The ignition delay time is indicated by dashed lines.

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 O₂-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 O₂, 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_{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

$$Nu = 2 + 0.552\sqrt{Re_p}Pr^{\frac{1}{3}}, \quad (4)$$

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, ρc_p) [4, 5, 13, 14], which is sufficiently larger for CO_2 compared to N_2 (e.g. a ratio $\text{CO}_2/\text{N}_2 = 1.7$ 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_{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 τ_i of a reactant gas mixture can be expressed as [48]:

$$\tau_i = \frac{c_v T_0^2 / T_a}{q_c Y_{F,0} A \exp(-T_a / T_0)}, \quad (5)$$

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.

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 C₃, C₄, C₇, and C₈ are included, and the corresponding t_{ign} are indicated by dashed lines in (b) and (c).

789 It is difficult to systematically explore the changes that CO₂ introduces by only
 790 exchanging the atmosphere, even under fully-defined conditions in the present ex-
 791 periments, since e.g. the flow rates for all CO₂ atmospheres are reduced to obtain
 792 gas temperature profiles similar to N₂ atmospheres. As a consequence, the parti-
 793 cle slip velocity is lower in OXY conditions, as shown in Fig. 12 which is another
 794 parameter crucially influencing the ignition and combustion behavior of solid fuel
 795 particles. With the aid of simulations, it has been discussed above that a higher slip
 796 velocity increases heat convection and hence shortens ignition delay if other boundary
 797 conditions remain unchanged. In other words, assuming the same slip velocity as in
 798 AIR conditions, t_{ign} in OXY conditions (see Fig. 14(b)) would further decrease lead-

ing to more significant differences between N_2 and CO_2 atmospheres. In summary it can be said, that an experimental study of the influence of CO_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_1 , C_2 , C_7 , and C_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_2 than in N_2 due to a higher c_v . For the same particle diameter, it can be seen that the local gas temperature $T_{g, local}$ is higher in the presence of CO_2 than within N_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_2 is about 1.2 times higher than that of N_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_2 atmosphere, which also implies an early formed flammable gas mixture. Equation 5 indicates that the gas temperature T_0 (here equivalent to $T_{g, local}$) and $Y_{F,0}$ have opposite effects compared with c_v or ρc_v on the ignition delay time. For example, Khatami et al. [15] explained in DTF measurements that the retarded ignition in CO_2 is mainly attributed to the lower ambient temperature compared to that in N_2 in their facility. Since differences of $T_{g, local}$ 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_2 with CO_2 does not necessarily result in a delayed ignition with the same initial global gas temperature. In general, CO_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 $T_{g,local}$ 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 C₁, C₂, C₇, and C₈ are compared, and the corresponding t_{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 .

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 $d_p = 120 \mu\text{m}$) and B (mean $d_p = 205 \mu\text{m}$) were investigated in four N_2 atmospheres (containing 10vol% - 40vol% O_2) and three CO_2 (containing 20vol% - 40vol% O_2) atmospheres. Multi-parameter optical measurements enabled the simultaneous detection of the ignition delay time t_{ign} and the volatile combustion duration t_{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 , $T_{g, \text{local}}$, $m_p/m_{p,0}$ and Y_F . By analyzing experimental and numerical observations and including theoretical evidences, the effects of particle size d_p , O_2 enrichment, slip velocity U_s , and CO_2 addition were discussed with regard to t_{ign} and t_{vol} . Several conclusions are summarized as follows:

- (1) Both t_{ign} and t_{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 $T_{g, \text{local}}$ 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_{vol} .
- (2) Both t_{ign} and t_{vol} decrease with an increasing O_2 concentration in all investigated atmospheres, with a more pronounced effect for larger particles. The higher partial pressure of O_2 accelerates the formation of a flammable gas mixture (i.e. shortens t_{ign}) and raises the fuel consumption rate (i.e. shortens t_{vol}) by faster molecular diffusion. For t_{vol} , the 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_{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_{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 .

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