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Optical diagnostics on coal ignition and gas-phase combustion in co-firing ammonia with pulverized coal on a two-stage flat flame burner

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Abstract

Gradual substitution of coal with green ammonia is a practical approach for the coal power phasedown at a minimal cost of modification, but the ignition and gas-phase reaction during co-firing NH₃ with coal remain largely unclear. In this work, we investigate the co-combustion behaviors of NH₃ and a high-volatile coal on a two-stage flat flame burner. Remarkably, the post-flame oxygen mole fraction $X_{i,02}$ of the inner stage can be manipulated to reproduce a proper reducing-to-oxidizing environment that coal particles experience in the practical combustor. We first reveal that, under certain values of $X_{i,O2}$ and NH₃ co-firing energy ratios $E_{\rm NH3}$, the reaction intensity (manifested by OH-PLIF signals) in the NH₃-coal flame is stronger than burning either pure coal or NH₃. This synergetic effect originates from an NH₃-combustion-induced enhancement of volatile release. We then propose a characteristic time scale τ_{OH} from the OH signals for the initiation of overall reactions in the system. In the case of $X_{i,O2}=0$, τ_{OH} monotonically increases with E_{NH3} , while for $X_{i,O2}=0.2$, the trend transitions to a decreasing one. It can be interpreted by comparing τ_{OH} with the characteristic O_2 diffusion time, coal particle heating time, and the coal pyrolysis time under different $X_{i,Q2}$. Furthermore, the coal particle ignition in coal-NH₃ flames can no longer be determined by visual images. Instead, we apply CH* chemiluminescence to identify the stages of coal particle ignition and volatile combustion in the NH₃-coal flame. While NH₃ addition has both positive (elevating temperatures & diluting coal particles) and negative (consuming O_2) effects on coal ignition, the combined influence of $E_{\rm NH3}$ is marginal on coal ignition delay time. On the other hand, the volatile combustion time decreases linearly with $E_{\rm NH3}$, suggesting a pure effect of reduced coal feed rate.

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1. Introduction

To limit global warming to 1.5°C, the *Glas-gow Climate Pact* calls for accelerating efforts towards the phasedown of unabated coal power [1].

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However, the economic growth of many developing countries is fueled by coal [2,3]. Coal is also a baseload power source to tackle the renewable energy intermittency [4]. Considering the vast installed capacity of coal power, replacing coal with carbon-free fuels at a minimal cost of modification is attractive for many countries.

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Ammonia (NH₃) is a promising alternative fuel in coal plants, with a low-heating value of 18.6 MJ/kg commensurate to low-rank coals [5]. Green NH₃ from renewables is projected to be a competitive, safe and long-duration energy/hydrogen carrier at scale [6]. Modern NH₃ trade via global transportation and the industrial use of NH₃ to reduce nitric oxides (NO_x) facilitate NH₃ as a fuel with existing infrastructure and regulations [5,7]. Thus, cofiring NH₃ with coal is a feasible approach for coal phasedown [8]. Several studies show that co-firing NH₃-coal achieves complete NH₃ burnout and that CO₂ reduction is in proportion to NH₃ co-firing ratios [9–13]. As for NO_x , it was demonstrated in MW-scale tests that below 20% NH₃ ratio of the input energy, NO_x emission is not exacerbated by injecting NH_3 into the fuel-rich zone [7,12,14]. These efforts preliminarily demonstrate the viability of co-firing NH₃-coal and motivate more ambitious goals. In particular, Japan's largest power generation company, JERA, plans to achieve a 20% cofiring rate of NH₃ in coal plants by 2030s and even 100% NH₃ replacement by 2040s [15].

However, fundamental investigations of NH₃coal co-combustion are limited. The existing studies mainly focused on measuring and modeling flame propagation of co-firing NH₃-coal. It was revealed that the spherical flame propagation speeds of NH₃-coal mixtures can be faster than burning pure coal or NH₃ by varying the equivalence ratio, fuel ratio, and turbulence intensity [16,17]. So far, little is known about the coal stream ignition and volatile combustion in co-firing NH₃ with coal, while these issues govern the flame stability of future coal-NH₃-fired boilers (with small to moderate NH₃ fractions) operated at flexible outputs. Coal ignition, on the other hand, was intensively studied on the ignition mode and ambiance effects [18–21]. Co-firing coal with NH₃ poses new challenges because coal ignition delay can no longer be quantified via visible-light intensities [20-22]. Instead, the approach needs to tell the two species apart. CH* chemiluminescence can be such an option [18,19,23]. Furthermore, OH-PLIF was widely used to characterize the reaction zones in coal [16,24-26] and NH₃ [27,28] combustion, and will be useful for NH₃-coal co-combustion studies.

We remark that the flat flame burner is suitable for the fundamental NH_3 -coal study with practical relevance. The existing MW-scale and boiler tests suggest injecting NH_3 from the burner center being preferable to side-wall injections because of the lower retrofitting cost and better performance of NO_x control [9,12,13]. In the case of central NH_3

Fig. 1. Schematic of the two-stage flat flame burner [29,30].

injection, coal particles mix with ammonia at a very early stage under local reducing and volatile-rich environments. The reducing-to-oxidizing environment that the coal-NH₃ mixture experiences can be well simulated on a novel two-stage flat flame burner with coal particles centrally entrained by NH₃ and N₂ [29,30]. Thanks to the ability of the burner to precisely adjust the ambience condition in a wide range [21], an investigation of co-firing NH₃-coal on the two-stage flat flame burner is of essence to promote the understanding of earlystage NH₃-coal interactions, but it is not reported yet.

In this paper, we successively study the ignition, volatile combustion, and reaction zone structure for co-firing NH_3 with coal on the novel two-stage flat flame burner under well-controlled temperatures, NH_3 ratios and local oxygen fractions. We have integrated visual images, CH^* chemiluminescence, and OH-PLIF to quantitatively characterize various time scales of NH_3 -coal co-combustion, and revealed a pattern transition of reaction initiation for different oxygen concentrations.

2. Experimental apparatus and methods

2.1. Two-stage flat flame burner and operating conditions

Fig. 1 shows the optically-accessible two-stage flat flame burner [29,30]. The burner consists of two regions (inner Φ 13 mm, outer Φ 80 mm) with hundreds of tiny non-premixed flames. By manipulating the flow rates of fuel (CO+less than 2% CH₄) and oxidizers (O₂+N₂), the post-flame temperature, O₂ fraction and gas velocity can be controlled separately.

In this work, the post-flame temperature is set at 1500 K in both regions and the outer O_2 mole fraction is fixed at 0.2. The inner O_2 mole fraction, $X_{i,O2}$, is varied as 0, 0.1, and 0.2 (denoted *case 1*, 2,

Case	$X_{i, O2}$	CO (SLM)	O ₂ (SLM)	N ₂ (SLM)
#1	0.0	0.34	0.15	0.99
#2	0.1	0.35	0.32	0.81
#3	0.2	0.36	0.54	0.73
outer region	0.2	6.63	12.77	29.64

Table 1 Gas flow rates in the investigated cases.

Fig. 2. (a) Post flame temperature (filled symbol) and oxygen mole fraction (hollow symbol) at the centerline for cases 1-3 without coal or NH₃ addition; (b-d) Post flame temperatures at the centerline with different NH₃ co-firing ratios $E_{\rm NH3}$ but no coal supply for (b) case 1, (c) case 2 and (d) case 3.

and 3). Table 1 gives the unburned inner flow rates in all cases, and the flow rates fed to the outer region are unchanged. Fig. 2 presents the measured profiles of gas-phase temperature and O_2 fraction (by the gas analyzer MRU-Vario Plus) at the centerline. The temperature was measured by a 0.5-mm bare B-type thermocouple (after radiation loss correction [31]).

Fig. 2a shows the temperature and oxygen fraction without addition of coal particles or NH₃. It verifies that the hot ambience is close to the designed temperature for all cases, and a process of O₂ diffusion inwards is observed in *cases 1&2* with smaller $X_{i,O2}$. In Fig. 2b-d, the centerline temperatures were measured in various $X_{i, O2}$ cases with NH₃ addition and no coal supply. For each case, the temperature increases with the ammonia cofiring ratio $E_{\rm NH3}$. In particular, the temperatures with $E_{\rm NH3} = 1.0$ can be up to 300K higher than those with $E_{\rm NH3} = 0$ (no central NH₃ fed in). Besides, with NH₃ addition, the larger- $X_{i,O2}$ cases reach temperature peaks at smaller heights above burner (HABs). In Fig. 2b ($X_{i,O2}=0$), it is seen that adding NH₃ results in a temperature drop close to the burner surface (HAB < 10 mm) for any $E_{\rm NH3}$ > 0. This is likely to be caused by NH₃ pyrolysis/ decomposition occurred under fuel-rich conditions at around 1200K [32].

To convert HAB *h* into the coal particle residence time $\tau(h)$, we used a Phantom v311 high-speed camera to measure the velocity and residence time of coal particles [30]. The recording speed was 3300 frames per second. More details are given in Section 2 of the supplementary material.

2.2. Fuel property

We used a high-volatile Zhundong lignite in this work, with the coal properties listed in Table 2. The pulverized coal sample was screened and dried before experiments. The coal feed rate was set at 0.08 g/min (\pm 5%) in cases burning pure coal. When NH₃ was added, the coal supply was reduced to keep a constant energy input. The NH₃ co-firing ratio (w.r.t. input energy) $E_{\rm NH3}$ was varied as 0, 0.2, 0.4, 0.6, 0.8 and 1.0 (pure NH₃ flame). Coal (with N₂) and NH₃ are fed in through the central tube of Φ 2 mm (see Fig. 1).

Proximate analysis (wt.%, dry basis)	
Fixed carbon	63.54
Volatile matter	30.58
Ash	5.88
HHV (MJ/kg)	28.83
<i>Ultimate analysis (wt.%, dry, ash-free basis)</i>	
С	71.60
Н	3.16
Ν	0.78
S _{total}	0.52
O (by difference)	23.94
<i>Vol. mean</i> d_p (μm)	28.39

Table 2Properties of Zhundong lignite.

Fig. 3. CH* chemiluminescence and OH-PLIF setups.

2.3. Optical diagnostics setup

Fig. 3 illustrates the optical setup for OH-PLIF and CH* measurements. For OH-PLIF measurements, the Q1(8) line of the OH $A^2\Sigma^+ \leftarrow X^2\Pi$ (1-0) transition band was used due to its low temperature sensitivity [33]. A 283 nm-UV laser beam (~8 mJ/pulse) generated by a Sirah Cobra-Stretch tunable dye laser (Rhodamine 590 dye) and a 5-Hz Nd:YAG laser were used to excite the Q1(8) line of the OH $A^2\Sigma^+ \leftarrow X^2\Pi$ (1-0) transition. The laser beam was tuned to a vertical sheet by a microcylindrical lens array (MCLQ(S)-509). The OH fluorescence at ~308 nm was recorded by an ICCD camera (Princeton PI-MAX IV, 1024 × 1024 pixels, gain 50, gate 55 ns) with 307 ±10 nm filters.

For CH* chemiluminescence, the signals were recorded by the ICCD camera (gain 50, gate width 5 ms) with an interference filter at 430 nm (10nm FWHM). The spatial resolution is 0.058 mm/pixel. Interference from thermal radiation was subtracted by the triple-filter technique (see Section 1 of the supplementary material) [18,34].

In each measurement, 100 images were acquired for OH-PLIF and 100 for CH*. Besides, the direct flame pictures were taken by a Nikon D5600 digital camera (2 ms exposure time, 4000×6000 pixels, 0.024 mm/pixel spatial resolution).

Fig. 4. Co-firing NH₃ with coal on the two-stage flat flame burner under various E_{NH3} and $X_{i,\text{O2}}$.

3. Results and discussions

3.1. Flame observation

Fig. 4 shows the visual images of NH₃-coal flames under varied values of $E_{\rm NH3}$ (from 0 to 1) and $X_{i,O2}$ (0, 0.1, 0.2). The orange pure NH₃ flame (a6, b6 & c6) can be stabilized on the flat flame burner under the high temperature (1500K) ambiance. Different from pure coal flame at the centerline, NH₃ diffusion flame forms a 'conical' surface. In some NH₃-coal flames, a dim NH₃ flame surface can be discerned upstream of the bright yellow coal flame. For any fixed $X_{i,O2}$, the flame brightness and 'coal flame length' both decrease with the increase of $E_{\rm NH3}$, while the position for 'coal particle ignition' seems to change marginally. When increasing $X_{i,O2}$ from 0 to 0.2, we see significantly brighter but shorter flames at a certain NH3 co-firing ratio, and the HABs where the flame intensity starts to surge substantially decrease for $E_{\rm NH3} \leq 0.8$.

We remark that unlike in pure coal combustion, the flame intensity can no longer be used as an indicator for coal ignition time when ammonia is added.

3.2. Coal ignition and volatile combustion

The CH* chemiluminescence was measured in this work to quantify the processes of coal ignition and volatile combustion in coal-NH₃ cocombustion. CH* is formed only from coal combustion. The early-stage coal devolatilization and volatile combustion are proved to generate large quantities of CH* radicals; Furthermore, char combustion also contributes to traces of CH* radicals [19,23]. Thus, CH* chemiluminescence not just distinguishes coal combustion from NH₃, but

Fig. 5. (a) Example of the spatial distribution of CH* signal after blackbody radiation correction (see Section 3.1 of the supplementary material for details). (b) CH* signal profile v.s. HAB and the quadratic function fit of the peak for deriving τ_i and τ_{vol} .

also helps decide ignition and volatile combustion stages in coal combustion.

Fig. 5a shows a typical time-averaged image of the spatial distribution of CH* chemiluminescence. Then, the signal intensities were averaged for each HAB within a width of 60 pixels around the centerline. The resulting profile, after a min-max normalization, is presented in Fig. 5b. The ignition delay time (τ_i) is defined as the onset of the CH^{*} signal, which indicates that the released volatile starts to oxidize [18,19]. The volatile combustion time (τ_{vol}) is defined as the duration time from the onset of ignition to the end of volatiles oxidation. In practice, these time scales are determined by the intersections of the baseline with a second-order polynomial used to fit the most significant (and the earliest) peak of the normalized CH* signal, as marked in Fig. 5b [19]. It is demonstrated that the secondorder polynomial fitting suffices to cover the sharp intensity rise as the particle ignited, and is less sensitive the non-uniform CH* emission toward the end of devolatilization [19]. Fig. S5 further verifies that τ_i is robust against the degree of the fitted polynomial. For each case, τ_i and τ_{vol} were measured three times, with 100 pictures recorded each time, and the relative errors are < 10%.

Fig. 6 presents τ_i and τ_{vol} in all cases with various $E_{\rm NH3}$ and $X_{i,O2}$. The coal ignition delay τ_i remarkably drops with the increment of $X_{i,O2}$ (~6 ms at $X_{i,O2}=0, \sim 3$ ms at $X_{i,O2}=0.1$, and ~1 ms at $X_{i,O2}=0.2$). Increasing $X_{i,O2}$ causes coal ignition to transition from a homogeneous mode (i.e., volatile ignites first) to a heterogeneous mode (i.e., surface ignition occurs first) [20–22]. The decline of τ_i between *cases 2* and 3 is milder than that between

Fig. 6. Derived τ_i (dotted line and hollow symbols) and τ_{vol} (solid line and filled symbols) under various E_{NH3} and $X_{i,O2}$. (*case 1*: $X_{i,O2}$ =0; *case 2*: $X_{i,O2}$ =0.1; *case 3*: $X_{i,O2}$ =0.2).

cases 1 and 2, implying a nonlinear dependence of τ_i on the oxygen fraction. The nonlinearity is also observed in the previous work on pure coal combustion [21].

The effect of NH₃ addition on τ_i is more complex. When E_{NH3} =0.2, a slight decrease in τ_i can be found especially for $X_{i,O2} = 0$ (*case 1*, ~1.2 ms) and 0.1 (*case 2*, ~ 1.2 ms). This may result from the NH₃-promoted homogeneous gas-phase ignition, denoted Route (*i*). For $X_{i,O2} = 0$, NH₃ pyrolysis dominates as revealed by the initial temperature drop in Fig. 2b. The formed H₂ (or H-containing species) accelerates the volatile reaction through both chemical and thermal effects. For $X_{i,O2} = 0.1$ and 0.2, NH₃ oxidation (combustion) emerges (see the temperature rise in Fig. 2c-d) and the thermal effect is the main reason for τ_i reduction.

However, with more NH₃ added, τ_i becomes only mildly affected by the NH₃ co-firing ratio. Notice that by Route (i), coal particles are heated up faster (denoted Route (ii)). Other possible interactions include: (iii) more dilute coal particles heat up and devolatilize more rapidly; (iv) the consumption of O₂ by NH₃ and the 'volatile barrier' [21] retard the surface reaction of coal. In low-oxygen cases 1 and 2, homogeneous ignition dominates. Route (i)plays the leading role with a small $E_{\rm NH3} < 0.2$. However, further increasing $E_{\rm NH3}$ seems to neutralize the positive effects on τ_i most likely due to Route (iv). Whereas, in case 3 where heterogeneous ignition prevails, increasing $E_{\rm NH3}$ to 0.8 even somehow delays coal ignition. We thus reasonably postulate that Route (*iv*) is responsible for the observed negative effects.

For volatile combustion, τ_{vol} exhibits a negative linear correlation with E_{NH3} in all cases. It might be interpreted by the reduced coal feed rate. As partly evidenced in Fig. 4, the coal flame shortens with increasing E_{NH3} . Then, with $X_{i,O2}$ increased from 0 (*case 1*) to 0.1 (*case 2*), τ_{vol} decreases by ~2

Fig. 7. Time-averaged (left) and single-shot (right) OH-PLIF images in all cases for different NH₃ blending ratios $E_{\rm NH3}$.

ms mainly because of the enhanced volatile oxidation. Further increasing $X_{i,O2}$ to 0.2 (*case 3*) leads to a marginal change of τ_{vol} . This can be attributed to a more intensive volatile release, as compared with *cases 1* and 2, after sufficient surface reaction, heat release, and particle heating up [22,35]. This offsets the effect of elevated oxygen fraction. It is noted that the prolonged volatile combustion time at higher oxygen fractions was observed in a previous study [18].

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3.3. OH-PLIF measurements of NH₃-coal flame

To study the reaction zone of NH₃-coal flames, Fig. 7 presents the spatial distribution of timeaveraged OH-PLIF signals (left) of 100 consecutive pictures with background noises removed and single-shot OH signals (right) for various $E_{\rm NH3}$ and $X_{i,O2}$. Due to the axisymmetricnon-premixed laminar jet flame, the OH distributions surrounding the unburnt fuel jet are coaxially symmetric.

In pure coal combustion (a6, b6 & c6), OH (as well as the radical pool) is mainly produced from volatile combustion [36,37]. Therefore, the emergence of OH signal can be viewed as a marker for (homogeneous) ignition. When $X_{i,02}=0$, the 'cone-shaped' central region without OH signal is due to the lack of O₂ diffused inwards (recall the O₂ profile in Fig. 2a). The 'OH hole' gradually disappears with $X_{i,02}=0.1$ and 0.2. With $E_{\rm NH3}$ increased from 0 to 0.2, the OH signal peak enhances for all values of $X_{i,02}$, and the region without OH inflates radially, indicating the radial diffusion of NH₃ and its consumption of O₂. Further increasing $E_{\rm NH3}$ from 0.2 to 1.0 changes the pattern of OH distribution gradually to that of pure NH₃ non-premixed flames.

It is worth noting that, for $E_{\rm NH3}$ ranging from 0.2 to 0.8, the area with strong OH signal intensities (say, the intensity > 400 a.u.) is apparently greater than those in pure coal and pure NH₃ flames, so too is the peak of OH signal, especially when $X_{i,O2} = 0.2$. Because OH is closely related to the reaction intensity, this phenomenon reveals a synergetic effect in co-firing NH₃ with the high-volatile coal under proper blending ratios: NH3 combustion heats the coal particle more rapidly and accelerates volatile release, which in turn strengthens the gas-phase reactions [24]. H₂-related species formed from NH3 decomposition may play important roles in the process. The reduced radiation heat loss in co-firing cases due to the weaker blackbody radiation of NH₃ flame, as compared with pure coal combustion, can be a reason as well. This effect is further confirmed by the gas-phase temperatures in Fig. 2b-d: the highest temperature with NH₃ but no coal always coincides with the greatest $E_{\rm NH3} = 1.0$, so there must be some NH₃-coal/ volatile interactions happened at certain $E_{\rm NH3}$. Let us remark that the synergetic effect of high-volatile coal-NH₃ is as well observed in the elevated turbulent flame speed of the fuel blends [24].

The frame-integrated OH-PLIF intensity provides a qualitative metric of combustion intensity

Fig. 8. Time-averaged axial (a-c) and radial (d-f) distributions of normalized OH signal (normalized by the OH signal of pure NH₃ flame E_{NH3} =1 in *case 1*, i.e. $X_{i,O2}$ =0).

[37-39]. For this purpose, we summed the timeaveraged OH signals in Fig. 7 both radially (for axial profiles) and axially (for radial profiles), and min-max normalized the curves by the maximal and minimal signals of pure NH₃ flame in case 1. The results are illustrated in Fig. 8. The radial profiles (Fig. 8d-f) of OH are symmetric around the center r=0, and increasing $X_{i,O2}$ from 0 to 0.2 moves the lateral peaks of signals to the centerline. As for the axial profiles (Fig. 8a-c), we see that the signal emerges, grows, peaks, and then diminishes. The peak generally increases with $E_{\rm NH3}$, indicating an improved combustion intensity. The signal peaks occur at residence times in the range of 17-20 ms for different values of $E_{\rm NH3}$ and $X_{i,O2}$.

We then extract a characteristic reaction initiation time τ_{OH} out of the axial profiles of OH signals in Fig. 8a-c. Analogous to τ_i from CH* chemiluminescence, τ_{OH} is defined in this paper as the residence time when the OH signal reaches 10% of the maximum value for each E_{NH3} and $X_{i,O2}$. So τ_{OH} indicates the initiation of intensive reactions of either coal or NH₃. Fig. 9 plots τ_{OH} for different E_{NH3} and $X_{i,O2}$. For pure coal combustion (E_{NH3} =0) in all cases, τ_{OH} is comparable to the ignition delay τ_i from CH* chemiluminescence (see Fig. 6) and decreases with a greater $X_{i,O2}$.

There is a remarkable pattern transition in Fig. 10 among *cases 1-3*. In particular, for $X_{i,O2}=0$ (*case 1*), τ_{OH} increases almost monotonically with $E_{\rm NH3}$ (~6.1 ms for $E_{\rm NH3} = 0$, and ~10.7 ms for $E_{\rm NH3} = 1$). On the contrary, for $X_{i,O2}=0.2$ (*case 3*), τ_{OH} decreases monotonically with $E_{\rm NH3}$ (~4.8 ms for $E_{\rm NH3} = 0$, and ~3.6 ms for $E_{\rm NH3} = 1$). For the intermediate *case 2* with $X_{i,O2}=0.1$, the reaction initiation time $\tau_{\rm OH}$ keeps almost constant (~5.3 ms)

Fig. 9. τ_{OH} derived from OH-PLIF signals in all cases for different values of E_{NH3} .

Fig. 10. A comparison among several important time scales in co-firing NH_3 with coal in the investigated cases.

within $E_{\rm NH3} \le 0.8$, and increases to ~ 7.5 ms for pure NH₃ combustion.

Let us remark the reliability of the τ_{OH} definition here. We claim that the phenomenon of pattern transition is 'robust' against other reasonable definitions of τ_{OH} with slightly different quantitative values. In supplementary material Fig. S6, τ_{OH} is taken as the time when the normalized intensity reaches 0.1 (rather than 10% of the peak in each curve). The same pattern transition is present therein as in Fig. 9.

3.4. Discussion: time scales in co-firing NH₃-coal

We need to figure out a theoretical interpretation of the pattern transition in Fig. 9 under different $X_{i,O2}$. For this purpose, a gas-phase flame-sheet model [21,40] incorporated with CPD (Chemical Percolation Devolatilization) [41] was used to simulate the temperature rise and devolatilization process of the coal particle under a hot (1500 K) ambiance. See details of the governing equations and model parameters specified in [21,40] and Section 4 of the supplementary material. With the most initial dynamic behaviors of coal particles solved, one then defines the characteristic particle heating time $\tau_{\rm h}$ as the time required for the particle temperature to reach $(1-e^{-1})$ of the final value, and the characteristic coal pyrolysis time τ_v as the time required for the released volatile matter to reach $(1-e^{-1})$ of the volatile matter content of the coal particle [21]. These time scales give insights into the dynamics of co-firing of coal-NH₃.

Fig. 10 presents a comparison among several time scales in all cases of NH₃-coal co-combustion. Note that the reaction initiation times for coal and NH₃ are taken from Fig. 9 as $\tau_{OH}(E_{NH3}=0)$ and $\tau_{OH}(E_{NH3}=1)$, respectively. In all cases, the simulated $\tau_{\rm h}$ and $\tau_{\rm v}$ are of the same order of magnitude. For $X_{i,O2}=0$, there is an additional time scale, $\tau_{\rm diff,O2}$, for O₂ diffusion from the outer region to the central coal-volatile-NH3 zone. From the measurement in Fig. 1, $\tau_{diff,O2}$ is estimated of the order of 10 ms, much longer than $\tau_{\rm h}$ and $\tau_{\rm v}$. Therefore, the reactions are diffusion-controlled, and the volatile is substantially released before oxidation. Since the pyrolysis products of Zhundong lignite, being light gases like H₂, CH₄ and CO [30], are more reactive than NH₃, adding NH₃ delays the reaction and increases τ_{OH} under $X_{i,O2}=0$, the homogeneousreaction-dominated regime. In contrast, the other limiting case $X_{i,O2}=0.2$ has no diffusion barrier, and the more reactive NH₃ (due to the higher $X_{i,O2}$) has a time scale even shorter than coal pyrolysis $\tau_{\rm v}$. Therefore, adding NH₃ promotes the initiation of reaction and reduces τ_{OH} under $X_{i,O2}=0.2$, in which the OH signal mainly comes from NH₃ oxidation. In the intermediate case $X_{i,\Omega_2}=0.1$, the initial OH signal comes from the reactions of both volatile and NH_3 .

4. Conclusions

This paper investigates coal ignition, volatile combustion, and reaction zone structures in cofiring NH₃ with a high-volatile pulverized coal on a two-stage flat flame burner. By manipulating the inner stage oxygen mole fraction $X_{i,O2} \le 0.2$, our experiments better mimic the atmospheres around coal particles in the practical furnace.

Resorting to CH^{*} chemiluminescence, we find that the coal ignition delay time is only mildly affected by the NH₃ co-firing ratio E_{NH3} : When E_{NH3} < 0.2, coal ignition is slightly promoted by the gas-phase NH₃ reaction (pyrolysis for $X_{i,O2}=0$ and combustion for $X_{i,O2}=0.1-0.2$); Further increasing E_{NH3} may offset the positive effect by consuming O₂. Volatile combustion time is linearly declined with E_{NH3} , which is a direct result of reduced coal feed rate.

We reveal a synergetic effect that enhances the early-stage reaction intensity of co-firing coal-NH₃. In particular, for E_{NH3} ranging from 0.2 to 0.8 of the blends, the area with strong OH signals, as well as the peak value, gets larger than those in pure coal and pure NH₃ flames. This results from strengthened gas-phase reactions of the NH₃-coal volatile mixture.

A characteristic time scale τ_{OH} from OH-PLIF signals is proposed to characterize the initiation of intensive reactions in the system. We reveal a pattern transition for E_{NH3} -dependence of τ_{OH} under different $X_{i,O2}$: In the diffusion-controlled homogeneous-dominated regime (low $X_{i,O2}$), NH₃ is less reactive than the volatile, so τ_{OH} increases with E_{NH3} ; In contrast, the high- $X_{i,O2}$ case features a faster NH₃ reaction than coal devolatilization, so τ_{OH} decreases with E_{NH3} .

The findings in this paper are beneficial for more in-depth investigations of coal-NH₃ reaction models, the NO_x issue and the coal particle size effect, which are our future work.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10. 1016/j.proci.2022.07.221.

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