[Title page]

1. **Effect of ammonia/oxygen/nitrogen equivalence ratio on spherical turbulent flame propagation of pulverized coal/ammonia co-combustion**

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Abstract

Because ammonia is one of the most promising candidates for energy carrier in the future, various applications of ammonia as a fuel are currently considered. One medium for utilizing ammonia is by introducing it to coal-fired boilers. To the best of our knowledge, this paper is the first to report the fundamental mechanism of the flame propagation phenomenon for pulverized coal/ammonia co-combustion. The effects of the equivalence ratio of the ammonia-oxidizer mixture on the flame propagation velocity of pulverized coal/ammonia co-combustion in turbulent fields were clarified by the experiments employing a unique fan-stirred constant volume chamber. The flame propagation velocities of pulverized coal/ammonia co-combustion, pure ammonia combustion, and pure pulverized coal combustion were compared. As expected, the flame propagation velocity of pulverized coal/ammonia was higher than that of the pure pulverized coal combustion for all conditions. However, the comparison of the flame propagation velocities of pulverized coal/ammonia co-combustion and that of the pure ammonia combustion, revealed that whether the flame propagation of the pulverized coal/ammonia was higher than that of the pure ammonia combustion was dependent on the equivalence ratio of the ammonia-oxidizer. This unique feature was explained by a mechanism including three competing effects proposed by the authors. In the ammonia lean condition, the positive effects, which are the strong radiation from the luminous flame and the increment of local equivalence ratio by the addition of volatile matter, are larger than the negative effect, which is the heat absorption by coal particles in preheat zone. In the ammonia rich condition, the effect of an increment of the local equivalence ratio by the addition of volatile matter turns into a negative effect. Consequently, the negative effects overcome the positive effect in the ammonia rich condition resulting in a lower flame propagation velocity of pulverized coal/ammonia co-combustion.

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Keywords: Coal combustion; Ammonia combustion; Co-combustion; Turbulence flame propagation; Spherical flame;

List of figure captions

1. Introduction

Coal is internationally recognized as an essential energy source because of its low cost due to the numerous worldwide reserves and stable supply. In most of coal-fired power plants, pulverized coal is used for combustion for electricity production. However, as concerns increase about adverse environmental effects of greenhouse gases (GHGs) emission because of power generation from precious fossil fuels, countries around the world have begun promoting the development of renewable and sustainable energy sources. Ammonia is one of the most promising candidates for a hydrogen energy carrier and is alternative to independent hydrocarbon-based fuels [1]. The merits of using ammonia as a fuel are many and important points of them are high hydrogen density, easy of liquifying and storing, synthesis from renewable energy sources, and being carbon-free [2,3]. However, low combustion intensity restricts its applications. Co-combustion is an attractive energy generating option from economic and environmental viewpoints. The feasibility of co-combustion of pulverized coal and ammonia was proved by researchers of the Central Research Institute of Electric Power Industry (CRIEPI) [4]. Furthermore, co-combustion of pulverized coal and ammonia has immense potential for reducing GHGs emission and improving energy security.

Moreover, a turbulence environment is used for pulverized coal combustion in most of the coal-fired power plants. Many experimental and numerical works related to turbulent pulverized coal combustion have been conducted. R. Kurose et al. and S.M. Hwang et al. studied the turbulent pulverized coal jet flame features [5,6]. K. Hadi et al. investigated the turbulence intensity and coal concentration effects on the spherical flame propagation of pulverized coal [7]. S. Ahn et al. explored combustion characteristics and flame structure of a turbulent pulverized coal flame employing a hydrogen pilot flame [8]. J. Hayashi et al. and H. Takahashi et al. studied the soot formation characteristics of pulverized-coal combustion by a turbulent jet flame [9,10]. The effect of turbulence on conservation rate of char particles was studied by J. Kruger et al. [11]. G.L. Tufano et al. studied the turbulence effect on the devolatilization and burning behavior of the coal particles [12]. While the utilization of ammonia in coal-fired boilers is a promising step in the direction of a carbon-free society, research focused on probing the turbulent co-combustion characteristics of pulverized coal and ammonia has not been much. Because flame propagation velocity was an important feature for characterization stability of the flame in a coal-fired burner [13], K. Hadi et al. conducted experiments to reveal the turbulent flame propagation behaviors of pulverized coal/ammonia co-combustion under a constant ammonia/oxidizer equivalence ratio [14]. In co-combustion, the ammonia/oxidizer flame played a key role in promoting coal flame propagation; however, the co-combustion characteristics in different ammonia/oxidizer equivalence ratios have not yet been reported. Therefore, fully understanding pulverized coal/ammonia co-combustion behaviors under different ammonia equivalence ratios is important for pulverized coal combustion fields. To the best of our knowledge, this is the first study to report the spherical turbulent flame propagation characteristics of pulverized coal/ammonia co-combustion under different ammonia/oxidizer equivalence ratios.

The present work aims to clarify the flame propagation behaviors of pulverized coal/ammonia co-combustion under different ammonia/oxygen/nitrogen equivalence ratios. Additionally, the influence of turbulence intensity was explored in the research. Based on the experimental results from present study, new models for pulverized coal particle clouds/ammonia co-combustion can be developed.

2. Experimental apparatus and methods

2.1 Experimental setup

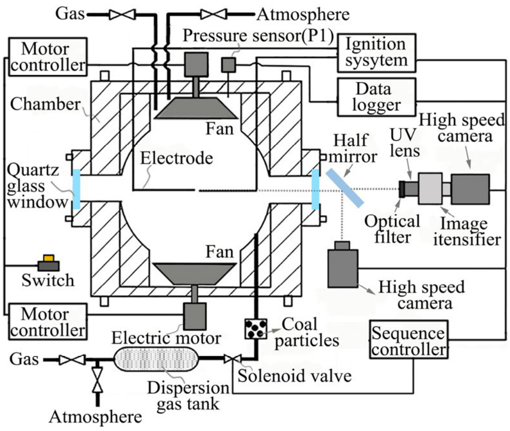

Fig. 1 Schematic of the experimental system

The schematic of the experimental setup is shown in Fig. 1. Experiments were carried out with a spherical chamber with a constant volume of 6.19×10^{-3} m^3 [15]. Moreover, a capacitor discharge ignition (CDI) circuit system was employed to ignite the mixture with an energy of 5.5 J [7].

Turbulence was generated by two identical counter-rotation seven-bladed fans that were vertically and symmetrically mounted inside the bomb. Turbulence parameters were measured by particle image velocimetry (PIV) measurements, as detailed in previous studies [7,15,16]. Within the central region of the vessel, the fact that the turbulence was homogeneous was established and no regular bulk motion existed [15]. Turbulence intensity, u', was proportional to the fan speed, f_s [16], found to be represented by Eq. (1):

$$u' = 0.00129 f_s. \quad (1)$$

Additionally, the integral length scale, L_f, was 20.9 mm regardless of the turbulence intensity. Five kinds of turbulence intensities were used for the research, including 0, 0.32, 0.65, 0.97, 1.29 m/s.

Four optical quartz glass windows, each with a diameter of 50 mm, were installed in the chamber. For the ammonia/oxygen/nitrogen flame, schlieren photography, which has been described in detail in previous research, was used to capture the flame propagation [15]. The resolution of schlieren photography was 896 × 896 at a frame rate of 2000–3000 fps. However, OH radical photography was selected to capture gas phase combustion in co-combustion because the flame image could not be obtained by schlieren photography. Furthermore, the luminous flame in co-combustion was obtained using a direct camera, which was the same approach previously used in pure coal combustion [7]. Additionally, the pressure inside the chamber during flame propagation was measured by a pressure sensor.

A half mirror was adopted for simultaneously recording the direct image and OH radical image from the same window. The direct image was observed by a high-speed camera (MotionPro X4) with a lens (Nikon, AI Nikkor 50 mm f/1.2S), and the OH radical image was taken by a high-speed camera (Phantom, Miro C210) in combination with the image intensifier (Hamamatsu, c9548-03) with a UV lens (Hamamatsu, 50 mm f/3.5) and a 300 nm bandwidth filter (Edmund optics, 67884 (Y158-23)) (The detailed information and the confirmation of reliability with the filter can be seen in Table A, Fig. A , and Fig. B in Appendix). To obtain high quality and enough images for analysis, the frame rates of the two high speed cameras was set as 2000 fps, and the pixels' settings for the direct camera and the OH radical photography were 512 × 512 and 896 × 896, respectively.

2.2 Experimental procedure

Common bituminous pulverized coal with a mean particle size of 48 μm was used for the experiments, and its properties have been listed in previous study [7]. To conduct a comparison with previous coal combustion research, the same diluted oxygen gases, which comprised 40 vol% O_2 and 60 vol% N_2, were used.

In case of co-combustion, ammonia and coal combustion simultaneously occurred in the chamber.

The thermal heat input from co-combustion was kept the same as that with pure coal combustion, which was under a coal concentration of 0.6 kg/m^3, by theoretically subtracting a portion of the coal particles' thermal heat input by ammonia combustion. Additionally, the total thermal heat input from co-combustion was 0.10325 MJ, which was calculated from the pure coal combustion by the lower heating value (LHV) of the bituminous coal of 27.8 MJ/kg. Table 1 shows the coal needed (by weight) in the mixing field on the condition of changing the ammonia equivalence ratio from 0.4 to 1.4.

Table 1
Coal needed in the calculation in co-combustion

Equivalence ratio of ammonia/oxygen/nitrogen in co-combustion filed	Thermal heat input from ammonia/oxygen /nitrogen combustion [MJ]	Coal needed [kg]
0.4	0.01539	0.00316
0.6	0.02122	0.00295
0.8	0.02618	0.00277
1.0	0.03045	0.00262
1.2	0.03416	0.00248
1.4	0.03742	0.00238

As the first procedure of experiments, the coal particles were equally filled into four filter cups connected to the inlets. Then, the combustion chamber and the dispersion tank were emptied to 0 kPa through a vacuum pump. Subsequently, the dispersion tank was filled with gases adhering to an equivalence ratio of NH_3-diluted oxygen (40%O_2, 60%N_2) at 300 kPa. Similarly, the gas mixture with the same composition was supplied into the chamber. However, the gas mixture in the chamber was set to 75 kPa for the purpose of igniting at atmosphere pressure after dispersion. The dispersion gas swept the coal particles into the center of the spherical field within 0.7 s. Thereafter, just 0.3 s after the dispersion ended, the mixture was ignited at 101 kPa (atmosphere pressure). In the turbulent experiments, the fan speeds were adjusted to give the desired turbulence intensity before the dispersion.

Same adjustment methods with previous studies were used [7,15]. The initial temperature of the mixture for all the experiments was 298 ± 5 K. The maximum errors of NH_3 concentration, O_2 concentration, pressure, and the coal concentration were 1%, 1.5%, 5%, and 3%, respectively. At least three experiments were conducted for each condition.

3. Results and discussions

3.1 Flame observation

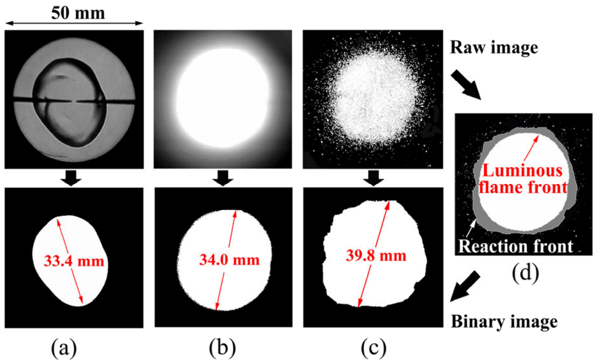

Fig. 2 Images processing; (a) ammonia/oxygen/nitrogen flame; (b) luminous flame in co-combustion; (c) OH radical image in co-combustion; (d) OH radical image processing

Figure 2(a) shows the schlieren image of ammonia/oxygen/nitrogen flames for ϕ = 0.8 at $u' = 0.32$ m/s . ϕ represents the ammonia/oxygen/nitrogen equivalence ratio of pure ammonia combustion. The 16-bit original schlieren image were processed by the grey level from 0 (black, background) to 255 (white, flame). Moreover, the threshold value was selected based on visual inspection. The threshold-value effect on the radius and velocity deviations were within ±3% (See Fig. C in Appendix). Fig. 2(b) shows the direct camera image of co-combustion on the condition of $\phi_{Ammonia}$ = 0.8 at $u' = 0.32$ m/s. $\phi_{Ammonia}$ represents the ammonia/oxygen/nitrogen equivalence ratio in co-combustion. The glow surrounding the luminous flame was not considered as part of luminous flame to maintain consistency with the pure coal combustion research that was previously conducted [7]. The luminous flame front comprises only the white border of the original image. When the number density of the soot particles, which are formed by the secondary pyrolysis of volatile matter evolved from coal particles, becomes sufficient, the luminous flame front was formed. Fig. 2(c) illustrates the OH radical image at same condition and time considered for Fig. 2(b). The border of the OH radical image is considered as the reaction front where the combustion reaction starts, which is the same as the consideration in many past combustion studies that have used fuels having hydrogen in its molecule. As shown in Fig. 2(d), the diameter of the reaction front (the border of OH radical image) is larger than the luminous flame front (the border of direct image). Finally, the flame radii were measured by counting the dimension between the farthest of the flame fronts, which was an approach that was similar to that used with pure coal combustion [7].

3.2 Flame propagation velocity

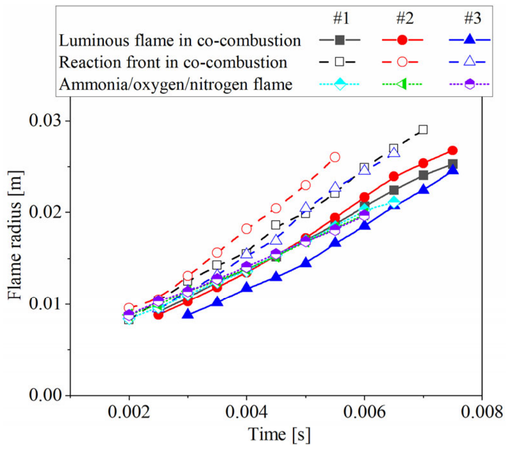

Fig. 3 Flame radii histories of the luminous flame and reaction front of co-combustion at $\phi_{\text{Ammonia}} = 0.8$ as well as ammonia/oxygen/nitrogen flame at $\phi = 0.8$ under the same turbulence intensity of $u' = 0.32$ m/s

Figure 3 shows the flame radii histories of the luminous flame, reaction front and ammonia/oxygen/nitrogen flame under the same equivalence ratio and turbulence intensity. Three trajectories of every flame in the same experimental condition are displayed here. The flame radius was measured from 0.008 m to the flame front touching the window edge for eliminating the ignition effect [16]. The pressure during observation range was constant at ±5% atmosphere pressure for all the conditions, which was a trend that was similar to previous studies [7,15,16]. As shown in Fig. 3, the luminous flame radius is always smaller than reaction front at the same time. This phenomenon was observed for all cases of co-combustion in the study. In addition, the reaction front radius was greater than that of ammonia/oxygen/nitrogen flame in this condition.

A polynomial relationship for the measured propagation radius as a function of time was applied to obtain the propagation velocity. For all turbulent combustion experiments, the propagation velocities of the luminous flame, reaction front and ammonia/oxygen/nitrogen flame increased with the increase of the flame radii, which was the same as that identified in previous pure coal combustion studies. Additionally, no constant value was observed within the observation range. Therefore, to examine turbulence intensity effect, the flame propagation velocities at a flame radius of 0.01045 m ($L_f/2$) were chosen for comparison [7]. Additionally, to compare the luminous flame propagation velocity of co-combustion with that of pure coal combustion and to compare the propagation velocities of the gas phase in co-combustion with that of ammonia/oxygen/nitrogen flame, the luminous flame, reaction front and ammonia/oxygen/nitrogen flame propagation velocities were separately analyzed and compared.

3.3 Luminous flame propagation characteristics in turbulent co-combustion under various ammonia equivalence ratios

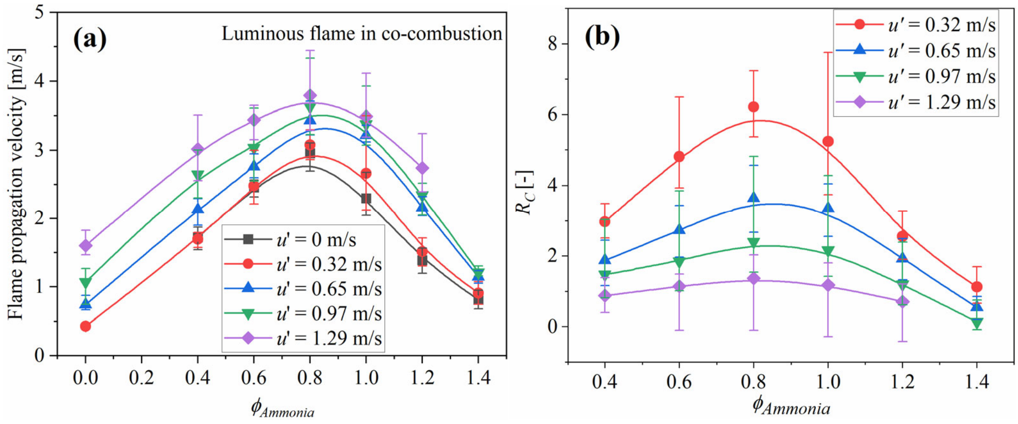

Fig. 4 (a) Luminous flame propagation velocities as functions of ϕ_{Ammonia} under different u'; (b) $Ratio_{Coal}$ in terms of ϕ_{Ammonia}

Figure 4(a) shows the luminous flame propagation velocities as functions of ϕ_{Ammonia} in co-combustion under various turbulence intensities. In addition, ϕ_{Ammonia} = 0 represents pure coal combustion [7]. As shown in this figure, the propagation velocity increases with the increase of turbulence intensity. This was considered to be caused by the enhanced turbulent transport of heat and mass of the pulverized coal particle clouds/ammonia/oxidizer mixture and the increased total surface area of the wrinkled flamelets by increasing turbulence intensity.

To examine the difference of luminous flame propagation velocities between co-combustion and pure coal combustion, $ratio_{Coal}$ (R_C) was used which was calculated by Eq. (2):

$$R_C = \frac{S_{LCO} - S_{LC}}{S_{LC}}, \qquad (2)$$

$S_{Luminous\ co\text{-}combustion\ flame}$ (S_{LCO}) represents the luminous flame propagation velocity in co-combustion. $S_{Luminous\ pure\ coal\ flame}$ (S_{LC}) represents the luminous flame propagation velocity of pure coal combustion, which is in same turbulent condition as with co-combustion. As shown in Fig. 4(b), all values of R_C

are positive. This means the luminous flame propagation velocity of co-combustion is larger than that of pure coal combustion. In addition, R_C increased with the increase of the ammonia equivalence ratio $\phi_{Ammonia}$ from 0.4 to 0.8. Then, a decreasing trend was observed with the increase of $\phi_{Ammonia}$ from 0.8 to 1.4. Vertically, under the same $\phi_{Ammonia}$, R_C decreased with the rise of the turbulence intensity.

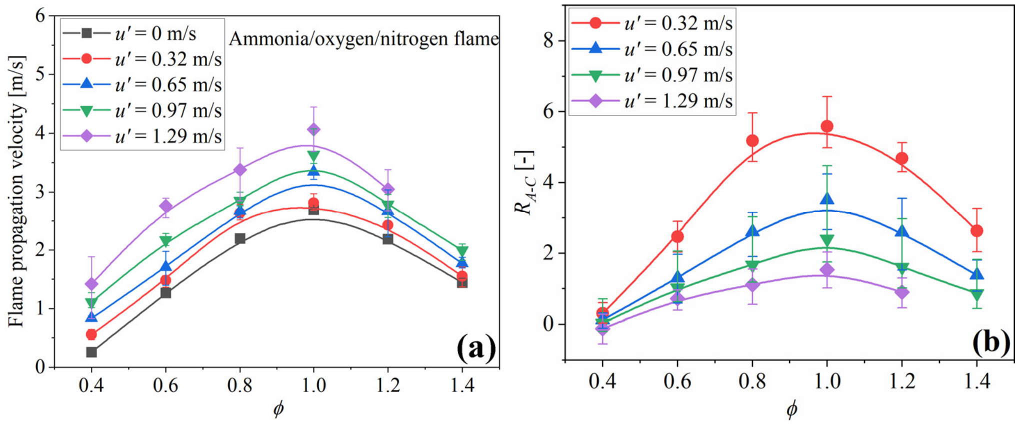

Fig. 5 (a) Ammonia/oxygen/nitrogen flame propagation velocities as functions of ϕ under different u'; (b) $Ratio_{Ammonia\text{-}Coal}$ as a function of ϕ under various u'

The disparity between propagation velocities of ammonia/oxygen/nitrogen flame with pure coal flame was examined by $ratio_{Ammonia\text{-}Coal}$ ($R_{A\text{-}C}$) that was expressed by Eq. (3):

$$R_{A\text{-}C} = \frac{S_A - S_{LC}}{S_{LC}}, \tag{3}$$

$S_{Ammonia/oxygen/nitrogen\ flame}$ (S_A) represents ammonia/oxygen/nitrogen flame propagation velocity, which is under the same turbulence intensity as with pure coal combustion. The relation of S_A as ϕ is shown in Fig. 5(a).

As shown in Fig. 5(b), all the values of $R_{A\text{-}C}$ under ϕ from 0.6 to 1.4 are positive, which is the same observed with R_C. Furthermore, under the same equivalence ratio, ϕ, $R_{A\text{-}C}$ decreased with an increase of the turbulence intensity, which was the same trend with R_C as $\phi_{Ammonia}$. Under the condition of $\phi_{Ammonia}$ from 0.4 to 1.2, the R_C at each $\phi_{Ammonia}$ is approximately the same with the $R_{A\text{-}C}$ at

$\phi = \phi_{Ammonia} + 0.2$ at the same turbulent condition. This was considered as the equivalence ratio increment by the volatile matter.

3.4 Reaction front propagation features in turbulent co-combustion under various ammonia equivalence ratios

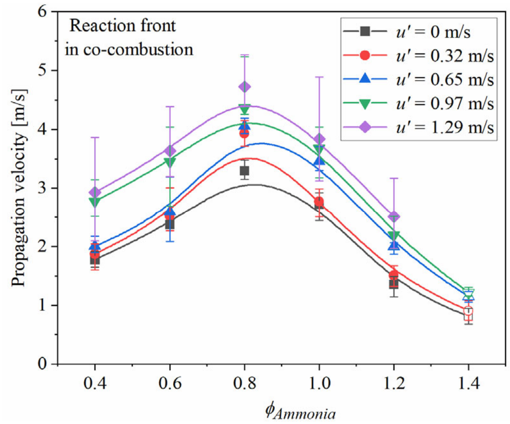

Fig. 6 Reaction front propagation velocities as $\phi_{Ammonia}$ under various u'

Figure 6 shows reaction front propagation velocities as functions of $\phi_{Ammonia}$ under various turbulence intensities. In the case of $\phi_{Ammonia} = 1.4$, the OH radical photography could not capture the OH radical because of weak light intensity. Therefore, for comparison with pure ammonia combustion, the propagation velocity of luminous flame front is used only for this condition ($\phi_{Ammonia} = 1.4$). This was because the propagation velocities of the reaction front in condition of $\phi_{Ammonia} = 0.4, 0.6, 1.0, 1.2$ at various turbulence intensities were approximately the same as those of the luminous flame. As shown in this figure, the propagation velocity increases with increase of the turbulence intensity, which is the same as the luminous flame propagation in co-combustion. By conducting a comparison with ammonia/oxygen/nitrogen combustion, the maximum propagation velocity for each turbulence intensity shifted to $\phi_{Ammonia} = 0.8$. This was because of the equivalence ratio increment by the volatile matter as explained in the last section. The released volatile matter increased the local equivalence ratio of the flame front.

To compare the propagation velocity of reaction front with that of ammonia/oxygen/nitrogen flame, $ratio_{Reaction\ front}$ (R_R) is used and is expressed by Eq. (4):

$$R_R = \frac{S_R - S_A}{S_A}, \tag{4}$$

$S_{Reaction\ front}$(S_R) represents the reaction front propagation velocity in co-combustion which is shown in Fig. 6. In this equation, the parameters were in the same turbulence intensity and ammonia equivalence ratio.

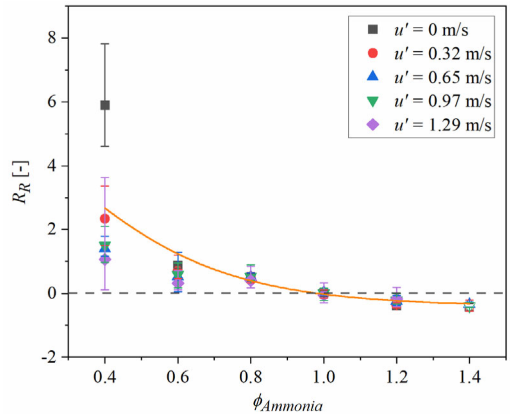

Fig. 7 $Ratio_{Reaction\ front}$ in terms of $\phi_{Ammonia}$ under various u'

As illustrated in Fig. 7, in ammonia lean conditions, the propagation velocity of the reaction front in the co-combustion case is greater than that of pure ammonia combustion. On the other hand, in ammonia rich conditions, the propagation velocity of reaction front in the co-combustion case is lower than that of pure ammonia combustion. However, on ammonia stoichiometric co-combustion, both are almost same. To explain this tendency, mechanism of pulverized coal/ammonia co-combustion has been proposed, as shown in Fig. 8.

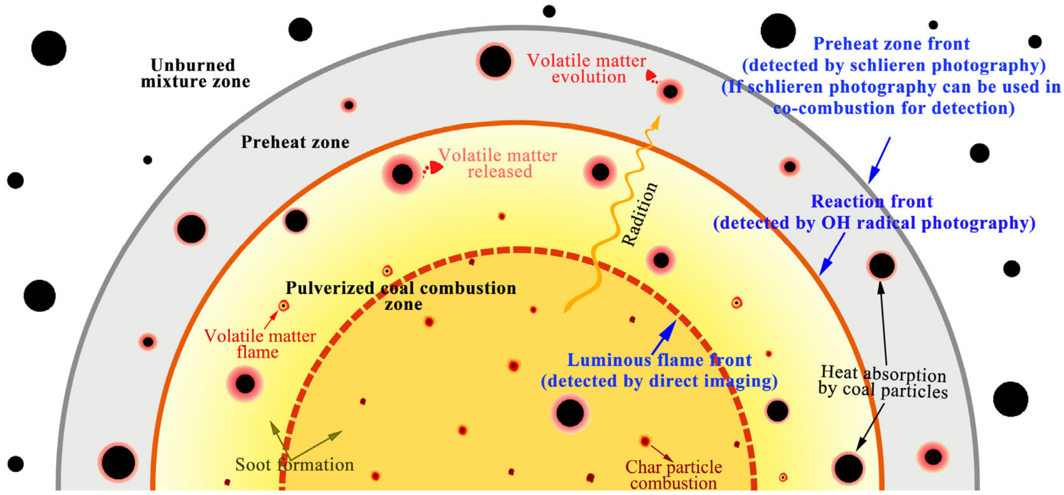

Fig. 8 One dimensional assumption of co-combustion flame structure

The pulverized coal/ammonia co-combustion flame can be divided into three zones according to the flame propagation direction, including the pulverized coal combustion zone, preheat zone, and unburned mixture zone. The reaction front is the border between the pulverized coal combustion zone and preheat zone. Most of the ammonia is considered to be completely combusted in a thin area of this reaction front in ammonia lean cases. In ammonia rich cases, most of oxygen is consumed by the reaction with ammonia and volatile matter in the reaction front. Some reactions including the devolatilization of coal particles and formation of soot particles proceed after the reaction front passing by. In the pulverized coal combustion zone, the single coal particle combustion follows the sequential process of heat-up, volatile matter release and combustion, and char combustion. Soot particles are formed by the secondary pyrolysis of volatile matter evolved from coal particles. Small diameter coal particles generally tend to combust faster than larger ones because of lager specific surface area [17]. Therefore, volatile matter release and combustion, soot formation, and char combustion simultaneously occur in this area. As defined previously, the luminous flame front is the white border of the original image, which indicates the existence of a high density of soot particles with high temperature. Furthermore, the luminous flame has a strong radiation effect to surrounding area [10].

In the preheat zone, coal particles with smaller diameter release volatile matter when the particles are heated up by the heat transfer from the reaction front and also by the heat transfer from the luminous flame in the pulverized coal combustion zone through the radiation. Before the reaction front passing by, the local equivalence ratio in the preheat zone was already changed by addition of volatile matter. Moreover, the unburned mixture zone is just a mixture of unburned gases and coal particles.

Clearly, the effects on the difference of flame propagation velocities between pulverized coal/ammonia co-combustion and pure ammonia combustion are of three types. First, the heat sink effect of the unburned coal particles in the preheat zone has a negative effect on the flame propagation velocity of the co-combustion. During the preheat of coal particles, the sensible heat as well as the heat required for the devolatilization are absorbed by the particles. Second, the strong radiation from the luminous flame to the unburned coal particles in the preheat zone has a positive effect on the flame propagation velocity of the co-combustion. This strong radiation is mainly caused by the soot particles with high temperature in the luminous flame [10] with a small contribution towards the radiation by the coal particles themselves. Indeed, the strong radiation between the solid particles in the pulverized coal combustion field is a unique additional heat transfer path [18], which is absent in the gaseous fuel combustion fields without soot formation. Third, the increment of local equivalence ratio in the preheat zone by addition of volatile matter can induce positive or negative effects, depending on $\phi_{Ammonia}$. This effect is positive in the ammonia lean condition because of the increase in the amount of fuel species that causes an increase in the flame temperature, while the increase in the amount in fuel has a negative effect in the ammonia rich condition because the addition of the fuel amount causes a decrease in the flame temperature. These three effects compete to determine the propagation velocity of the reaction front in co-combustion.

In the ammonia lean co-combustion condition, the propagation velocity of reaction front is larger than that of pure ammonia combustion. This is because the positive effects, which are the strong radiation from the luminous flame and the increment of local equivalence ratio by the addition of volatile matter, are larger than negative effect, which is the heat absorption by the coal particles. On the other hand, in ammonia rich condition, the negative effects, which are the heat absorption by coal particles and a very high equivalence ratio under a rich $\phi_{Ammonia}$ condition, overcome the positive effect of the strong radiation. In the ammonia stoichiometric condition, the propagation velocity of the reaction front is almost the same as that of the ammonia/oxygen/nitrogen flame. This is because the negative effect is offset by the positive effect.

The present research revealed the fundamental mechanism of the pulverized coal particle clouds/ammonia co-combustion. From a micro perspective, the single coal particle combustion phenomenon differs under conditions of different particle size, atmosphere gases composition, and ambient temperature [19–21]. The unique phenomenon of the single coal particle/ammonia co-combustion is considered to contribute to reveal the mechanism of pulverized coal/ammonia co-combustion. However, it is beyond the scope of the present study.

4. Conclusions

The effect of the ammonia/oxygen/nitrogen equivalence ratio on the flame propagation characteristics of pulverized coal/ammonia co-combustion under various turbulence intensities were investigated in this work. The principal findings are mentioned below.

The flame propagation velocity of pulverized coal/ammonia co-combustion is higher than that of pure ammonia combustion in the ammonia lean condition. Conversely, the flame propagation velocity of pulverized coal/ammonia co-combustion is lower than that of pure ammonia combustion in the ammonia rich condition. In the stoichiometric condition, the flame propagation velocities between co-combustion and pure ammonia combustion are approximately the same. The mechanism including three types of effects was suggested to explain the above results. In the ammonia lean condition, the positive effects, which are the strong radiation from the luminous flame and the increment of local equivalence ratio by the addition of volatile matter, are larger than the negative effect, which is the heat absorption by coal particles in the preheat zone. In the ammonia rich condition, the effect of increment of the local equivalence ratio by the addition of volatile matter becomes negative. Consequently, the negative effects overcome the positive effect in the ammonia rich condition resulting in the lower flame propagation velocity of pulverized coal/ammonia co-combustion compared with that of pure ammonia combustion. In the ammonia stoichiometric condition, the positive and the negative effects are comparable resulting in a flame propagation velocity that is almost same between co-combustion and pure ammonia combustion.

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