



Additional criteria for MILD coal combustion

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Received 7 November 2019; accepted 7 June 2020

Available online xxx

Abstract

We proposed a theoretical basis for Moderate or Intense Low-oxygen Dilution (MILD) coal combustion based on the turbulent scalar energy spectra. This is motivated by the hypothesis that smallest scalar mixing length scales should be on the order of the particle size or smaller to ensure that mixing can occur to prevent formation of diffusion flames. Our proposed criterion is evaluated using several experimental datasets from the literature for coal combustion in both MILD and traditional combustion regimes. The experimental results confirm that the smallest mixing length scales should be of the order of or smaller than the particle diameter, $\eta_{\text{mix}} \lesssim d_p$, to breakup the heat and mass transfer boundary layers around particles in MILD coal combustion. Results indicate that poor mixing of species with small Schmidt numbers around small particles leads to the high luminous intensity in the reactor. The effects of inlet velocity and jet diameter on the mixing length scales are analyzed. Higher inlet velocity and smaller jet diameter are expected to reach MILD regime. The proposed criterion can be used to guide experimental design to achieve MILD conditions for coal combustion.

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Keywords: MILD; Coal combustion; Particles; Scalar spectra

1. Introduction

Moderate or Intense Low-oxygen Dilution (MILD) combustion uses product recirculation to dilute the reactants prior to combustion. This dilution reduces the oxidizer concentration which leads to lower chemical reaction rate and a volumetric reaction zone, eliminating visible flames. To avoid formation of flames and instead favor volumetric combustion, MILD relies on fast mixing (dilution)

of products with the reactants and a mixture temperature that exceeds the autoignition temperature.

While MILD combustion of gaseous fuels has been studied fairly widely (see, *e.g.*, [1–8]), MILD combustion using solid fuel such as pulverized coal particles has received less attention. Inlet parameters, including the premixing patterns [9], burner configuration [10] and Reynolds number [11], have a significant influence on the formation of recirculation zone of the flue gas and the scalar mixing degree in the reactor. Saha, et al. [12,13] reported that the particle size and coal type have a big impact on the occurrence of devolatilization and the NO_x emissions on MILD combustion. These inves-

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<https://doi.org/10.1016/j.proci.2020.06.175>

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tigations indicated that sufficient mixing between oxidizer, recirculated flue gas and volatiles from coal particles before ignition are important on the achievement of MILD combustion. The investigations about the impacts of carrier gas [14,15], concentration of O_2 in the oxidizer [16] and H_2O additions [17] indicated the importance of sufficient dilution of oxygen to achieve MILD combustion.

Criteria for MILD combustion developed for gaseous fuels require that the inlet temperature of the reactant mixture is higher than the mixture self-ignition temperature while the maximum temperature increase with respect to the inlet temperature in the reactor should be lower than the mixture self-ignition temperature [18]. While the work summarized above emphasizes the importance of both turbulent mixing and chemical reaction to achieve MILD regime for pulverized coal combustion, there has not been any theoretical guidance on how to achieve MILD coal combustion.

In pulverized coal combustion, the fuel is introduced to the reactor as particles release volatiles at length scales on the order of the particle diameter. To obtain well-mixed reactants around the particles, criteria involving particle length scales relative to turbulent mixing scales seem appropriate. The main purpose of this work is to propose a criterion for MILD coal combustion that considers the mixing environment at the particle length scale. To accomplish this, we analyze the scalar turbulent mixing process using scalar energy spectra to relate the particle size and mixing length scales. We verify our proposed criterion using available literature data where MILD and traditional combustion regimes were reported for coal combustion.

We restrict ourselves to volatiles combustion which is a gas-phase phenomena and do not consider char oxidation/gasification since that is a surface phenomena and therefore cannot be volumetric by definition. However, we note that products of char gasification (predominantly syngas) can subsequently react in the gas phase; our analysis should apply there as well.

2. Mixing process for MILD coal combustion

The most important factor to reach MILD regime is the recirculation of flue gases. The recirculated flue gas dilutes the oxidizer and preheats the reactants. Mixing the reactants and recirculated products rapidly avoids the formation of a flame front. The low oxidizer concentration in the diluted reactants reduces the peak temperature and chemical reaction rates in the reactor. The mixing rates of reactants are always bigger than the reduced chemical reaction rates to avoid the formation of diffusion flames, and the elevated temperature results in mixtures exceeding the autoignition temperature thereby preventing formation of premixed flames.

In coal combustion, fuel is delivered to the gas phase as particles heat up. This means that the mixing environment at the particle length scale is important to mix the fuel with the oxidizer and recirculated product stream. To get well-mixed reactants around these particles, the local turbulent mixing environment should be small enough to breakup the heat and mass transfer boundary layers that form around particles. To analyze this, we consider homogeneous turbulence, where well-developed theory describes the turbulent mixing structures through energy spectra. By tying this to system-level parameters (Reynolds number and particle size), we relate the particle size to local mixing length scales.

The separation of the Kolmogorov (η) and integral (L) length scales is related to the Reynolds number as

$$\eta/L = Re_L^{-3/4}, \quad (1)$$

where Re_L is Reynolds number based on integral length scale [19]. Small-scale scalar structures are affected by not only Re but also Schmidt number, $Sc \equiv \nu/D$, where ν is the kinematic viscosity of the fluid and D is the mass diffusivity [20]. The scalar spectra is different for $Sc \leq 1$, and $Sc > 1$ and we briefly review this in Sections 2.1 and 2.2.

2.1. Scalar spectra for $Sc \leq 1$

For $Sc \leq 1$, the Obukhov–Corrsin length scale (η_C) is larger than the Kolmogorov scale, and the two are related by [21,22]

$$\eta_C = \eta Sc^{-3/4}. \quad (2)$$

Inertial-convective range. The classical Kolmogorov–Richardson cascade [23] yields the scalar spectra $E_\phi(\kappa)$ for the wavenumbers κ in $1/L < \kappa < 1/\eta_C$,

$$E_\phi(\kappa) = C_{OC} \langle \chi \rangle \langle \epsilon \rangle^{-1/3} \kappa^{-5/3}. \quad (3)$$

Here, $C_{OC} = 0.5$ is the Obukhov–Corrsin constant, $\langle \chi \rangle$ is the mean scalar dissipation rate, and $\langle \epsilon \rangle = \nu^3/\eta^4$ is the mean kinetic energy dissipation rate. Because $\langle \chi \rangle$ only scales E_ϕ and doesn't affect L/η , we choose $\langle \chi \rangle = \langle \epsilon \rangle$.

Inertial-diffusive range. The scalar spectra for $\kappa > 1/\eta_C$ is given as [24]:

$$E_\phi(\kappa) = C_{OC} \langle \chi \rangle \langle \epsilon \rangle^{-1/3} \kappa^{-5/3} \exp\left(-\frac{3}{2} C_{OC} (\kappa \eta_C)^{4/3}\right). \quad (4)$$

2.2. Scalar spectra for $Sc > 1$

For $Sc > 1$, the Batchelor scale (η_B) is smaller than the Kolmogorov scale, with the two related by [25]

$$\eta_B = \eta Sc^{-1/2}, \quad (5)$$

Inertial range. The Kolmogorov–Richardson cascade [23] yields the same scalar spectra $E_\phi(\kappa)$ for $1/L < \kappa < 1/\eta$, given by (3).

Viscous-convective range. The scalar spectra for $1/\eta < \kappa < 1/\eta_B$ is given as [20,24]:

$$E_\phi(\kappa) = C_B \langle \chi \rangle \left(\frac{\nu}{\langle \epsilon \rangle} \right)^{1/2} \kappa^{-1}. \quad (6)$$

where $C_B \approx 2$ is the Batchelor constant [26].

Viscous-diffusive range. The scalar spectra for $\kappa > 1/\eta_B$ is given as [20,24]:

$$E_\phi(\kappa) = C_B \langle \chi \rangle \left(\frac{\nu}{\langle \epsilon \rangle} \right)^{1/2} \kappa^{-1} \exp(-C_B(\kappa\eta_B)^2). \quad (7)$$

2.3. Particle sizes in the scalar spectra

The particle size relative to η_C and η_B determines the mixing environment for species with $Sc \leq 1$ and $Sc > 1$ respectively. Figure 1 shows the spectra discussed in Sections 2.1 and 2.2 as a function of wavenumber for various Re (Fig. 1(a)) and Sc (Fig. 1(b)). The vertical dashed lines in these figures represent smallest scalar mixing length scale, $\eta_{mix} = \min(\eta_B, \eta_c)$.

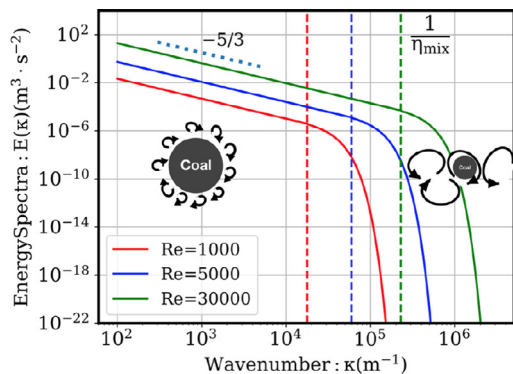
Figure 1(a) shows the effect of Re for $Sc = 1$. Fuel-containing particles can be placed on the x-axis at wavenumber $\kappa_p = 1/d_p$. If the smallest mixing length scale is smaller than the particle size, $\eta_{mix} = \min(\eta_B, \eta_c) < d_p$, then the particle sits to the left of the vertical dashed lines (representing the mixing length scale η_{mix}). In this case, all scalars will be mixed by turbulent eddies near the particle surface and boundary layer formation will be impeded. Therefore, well-mixed reactants should be obtained and MILD combustion should be possible.

On the other hand, if the smallest mixing length scale is bigger than the particle size ($\eta_{mix} > d_p$), then the particle sits to the right of the vertical dashed lines. In this situation, the turbulent scalar structures are larger than the particles and scalar boundary layers will form. The formation of these boundary layers implies a diffusive mixing between fuel and oxidizer/products which may result in nonpremixed flames. Achievement of MILD combustion under this condition depends on the local mixing and reaction time scales.

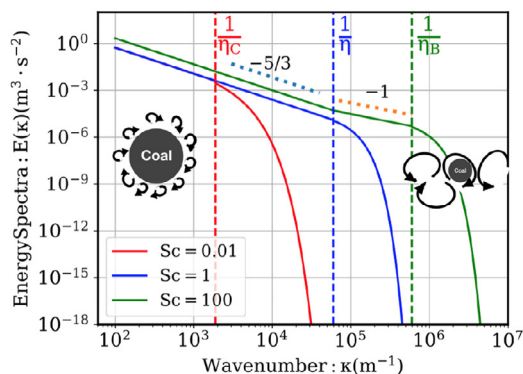
Therefore, to get well-mixed reactants around particles for MILD coal combustion, the smallest relevant scalar length scale (Batchelor or the Obukhov–Corrsin, depending upon Sc), should be smaller than the particle size,

$$\eta_{mix} = \min(\eta_B, \eta_c) < d_p. \quad (8)$$

with η_C and η_B given by (2) and (5), respectively. Eq. (8) is our proposed criterion for MILD coal combustion. This criterion provides *a priori* guidance for the setup of experimental conditions since, for a given particle size and integral length scale, it



(a) Different Re with $Sc = 1$.



(b) Different Sc with $Re = 5000$.

Fig. 1. Scalar energy spectra for different Re and Sc. Vertical dashed lines represent the smallest mixing length scales. The dotted lines labeled $-5/3$ and -1 represent the exponents of the wavenumber in Eqs. (3)–(4) and (6)–(7) to calculate scalar energy spectra.

can provide a required Re to achieve MILD. Alternatively, for a specific Re, it can provide the minimum particle size which can achieve MILD.

Figure 1(b) shows the scalar spectra at $Re = 5000$ for $Sc = [0.01, 1, 100]$ and illustrates the significant impact that Sc can have. Many of the volatile species evolving from coal have Sc near unity, but $Sc_{CH_2} \ll 1$. One could apply the criteria given by (8) to all gas phase species, but this may not be strictly necessary. Rather, there likely exist some subset of species for which this criterion applies. However, fuel components giving rise to the most significant heat release should, at a minimum, be included in this criteria. Furthermore, these fuel species are the ones primarily responsible for boundary layer formation around the particles, which further suggests their use in defining the range of Sc to consider. Identifying

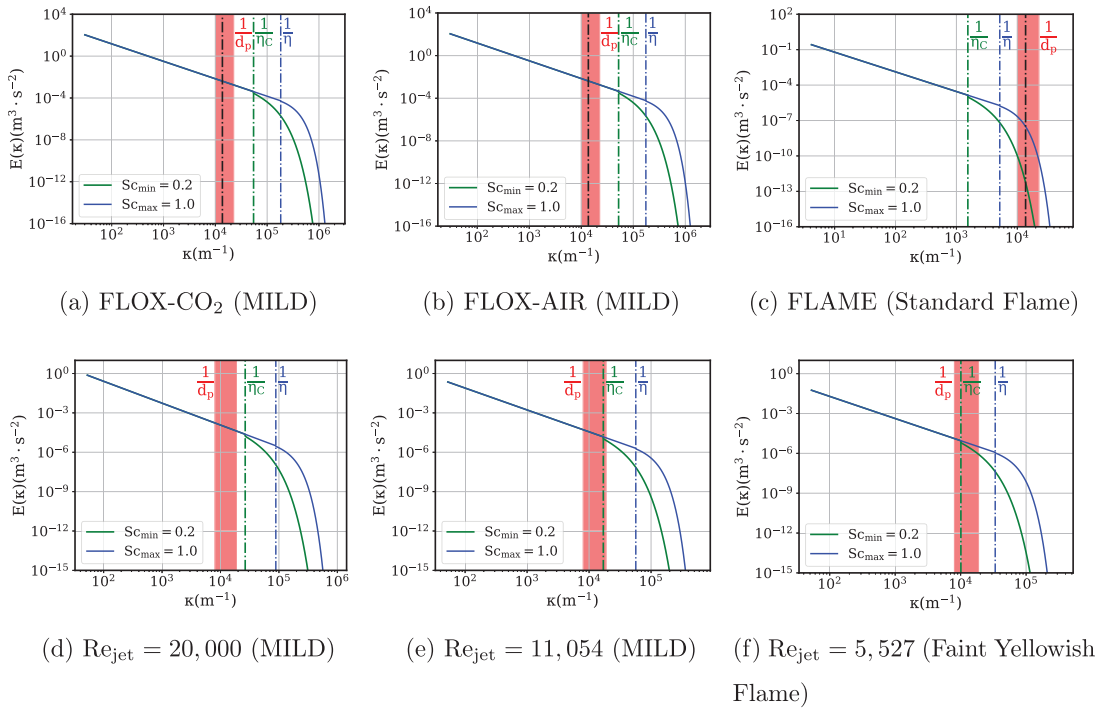


Fig. 2. Particle position in scalar spectra from literature data. Figure 2(a)–(c) use data from [14] while Fig. 2(d)–(f) use data from [11]. Blue and green dashed lines represent smallest mixing length scales based on max/min species Sc numbers. The red zone represents the range of particle sizes. The black dashed line in the red zone in Fig. 2(a)–(c) represents the mean particle size.

which species should be included in this criteria (specifically if low- Sc species such as H_2 should be included) should be a subject of future research.

In summary, to get well-mixed reactants around coal particles, a criterion relating particle size to the turbulent mixing is required. The smallest mixing length scale, varying with Sc , needs to be smaller than the particle size. This is expressed mathematically by (8). This criterion is valuable since it allows us to use system-level parameters (Re , L and d_p) together with fluid properties (Sc) to identify operating conditions that *may* be MILD to guide experimental design. Eq. (8) provides an additional criterion for MILD coal combustion with a specific particle size. For a particle size distribution, the criterion could be convoluted with a function that indicates the importance that particles of a specific size play in determining the combustion regime. MILD combustion might not be achieved if small particles with, $d_p < \eta_{mix}$, play significant roles in distinguishing the combustion regime. Eq. (8) is a necessary but not sufficient condition for MILD coal combustion; recirculation ratios are also a key parameter affecting MILD as discussed in Section 1. However, this additional

criterion applies to coal systems where fuel is delivered to the gas as particles devolatilize.

3. Results and discussion

3.1. Verification of mixing criteria for MILD coal combustion

To test our proposed criterion given by (8), we consider data from the literature where MILD as well as traditional coal combustion are reported. The smallest scalar mixing length scales for different species in the reactor are calculated using reported information for each case based on (8), which are compared with the particle size. The main parameters required from the literature are the inlet Re , integral length scale L , temperature and composition of the inlet streams and coal analysis (to obtain estimates of Sc), and the coal particle size distribution. The inlet Re is calculated using the jet diameter and gas parameters through the jet. For the cases where there are various jets for primary and secondary jets, area-weighted parameters, namely characteristic velocity u and integral length scale L , are used to calculate Re and mixing

Table 1
Mixing length scale analysis of literature data.

Ref.	Case	Re _L	d_p ($\bar{d}_p$) (μm)	η_{mix} (μm)	
				η	η_C
[14]	FLOX-CO ₂	110,000	44–100	5.5	18
	FLOX-AIR	100,000	(73.1)	5.8	19
	FLAME	13,000		190	650
[28]	CO ₂ -Carrier	23,000	38–180	3.1	11
	N ₂ -Carrier	18,000		3.6	12
[11]	Case 1	20,000	53–	11	38
	Case 2	11,054	125	18	59
	Case 3	5527		30	100
[13]	Case 1	20,000	53–125	11	38
	Case 2		250–355		
[29]	High T_{Air}	2600	(30)	41	130
	Low T_{Air}	6500		21	70

length scales,

$$\phi = \frac{N_{\text{prim}} D_{\text{prim}}^2 \phi_{\text{prim}} + N_{\text{sec}} D_{\text{sec}}^2 \phi_{\text{sec}}}{N_{\text{prim}} D_{\text{prim}}^2 + N_{\text{sec}} D_{\text{sec}}^2}, \quad (9)$$

where ϕ represents characteristic velocity u , or integral length scale L , N is the number of jets with diameter D . $N_{\text{prim}} D_{\text{prim}}^2$ and $N_{\text{sec}} D_{\text{sec}}^2$ represent the weights of parameter ϕ (u or L) from primary and secondary jets, respectively. The subscripts ‘prim’ and ‘sec’ indicate the primary and secondary jets, respectively.

We consider volatiles consisting of CO₂, CO, H₂O, CH₄, H₂, HCN, NH₃, consistent with the CPD model [27]. The composition and temperature of primary and secondary jets in the experiment, weighted by (9) are used to calculate Sc for each of the volatile species. The max/min Sc among these species in all experimental cases considered gives $\text{Sc}_{\text{max}} = \text{Sc}_{\text{CO}_2} \approx 1.0$ and $\text{Sc}_{\text{min}} = \text{Sc}_{\text{H}_2} \approx 0.2$, which is the Sc range used in the remainder of this paper.

Figure 2 shows the scalar spectra for six experiments [11,14] for illustrative purposes. The mixing length scales (blue and green dashed lines), representing the range of the smallest mixing length scale for different species in the reactor, are compared with the range of particle size (red zone) and mean particle size (black dashed line) in the spectra. Comparing the mixing length scale η_{mix} with the particle size distribution in Fig. 2(a) and (b), we see that $\eta_{\text{mix}} < d_p$ for the full particle size range. These cases both correspond to situations where MILD combustion was observed [14]. On the other hand, Fig. 2(c) shows that $\eta_{\text{mix}} > d_p$ for all particle sizes, and corresponds to a case where traditional combustion (with visible flames, see Fig. 4(b)) was observed [14]. These observations are consistent with the theoretical condition we propose in (8).

Figure 2(d) and (e) shows situations where the experiments indicate that these were MILD combustion [11]. The proposed criteria (8) is satisfied

in Fig. 2(d), but it is satisfied for only *part* of the particle size range for species with the lowest Sc in Fig. 2(e). For species with $\text{Sc} \geq 1$ and for the largest particles, the proposed MILD criteria is satisfied. The apparent inconsistency for small particles and species with $\text{Sc} < 1$ may be due to the Sc calculation including species such as H₂ that are highly diffusive but may not directly influence MILD. It could also be that the smaller particles are clustered together and act like larger particles so that MILD is still achieved. Definitely addressing this will require additional experiments and likely high-fidelity simulations. It is interesting to note that the experiments reported that higher luminous intensity for the $\text{Re}_{\text{jet}} = 11,054$ case (Fig. 2(e)) than the $\text{Re}_{\text{jet}} = 20,000$ case (Fig. 2(d)) as shown in Fig. 4(a) [11]. This is consistent with our analysis which suggests that the mixing near the particles degrades for some species for the $\text{Re}_{\text{jet}} = 11,054$ case.

Figure 2(f) shows a case where the experiments report faint, yellowish flames (see Fig. 4(a)) [11] suggesting that the experiment was bordering MILD/traditional combustion. Our analysis in Fig. 2(f) is consistent with this observation, indicating that only species with $\text{Sc} \geq 1$ are in the MILD regime over all particle sizes.

The results from Fig. 2 for two different experimental datasets observing MILD combustion suggest that our proposed criteria does, indeed, allow us to discriminate between MILD and traditional combustion.

Table 1 summarizes the findings from Fig. 2 as well as results from several other experiments available in the literature. The turbulent jet Re, Re_L, particle size (mean particle size), d_p ($\bar{d}_p$), and mixing length scales, η_{mix} (for $\text{Sc} = 1.0$ and $\text{Sc} = 0.2$) for each case are listed in the table. MILD combustion is reported in all cases except for the cases highlighting in gray, where a standard flame is observed. The key point of comparison in Table 1 is the particle size (d_p) relative to the smallest scalar

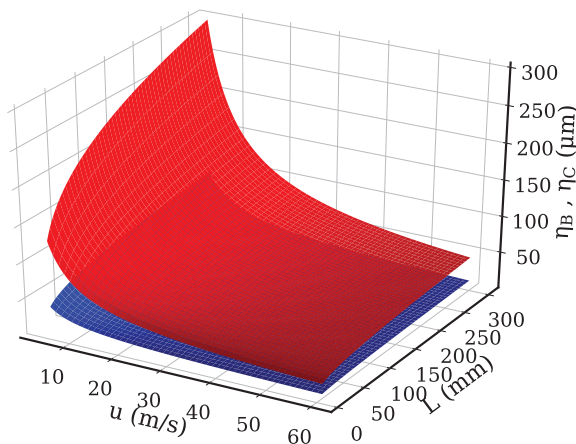


Fig. 3. Effects of inlet velocity (u) and integral length scale (L) on the mixing length scales with $\nu = 3 \times 10^{-5} \text{ m}^2/\text{s}$. The top red surface represents the mixing length scale for $Sc = 0.2$, above which MILD is expected. The bottom blue surface represents the mixing length scale for $Sc = 1.0$, below which traditional combustion is expected.

mixing scale η_{mix} . Specifically, note that in each case where MILD is reported experimentally, $\eta \ll d_p$. For $Sc < 1$, $\eta_C < d_{p,\text{min}}$ is observed in all MILD cases except for ‘Case 2’ from [11]. As discussed previously, flame luminous intensity is higher in this case because of the poor mixing of some species around small particles. For ‘Case 3’ in [11] which is bordering MILD/traditional combustion, $\eta < d_p$ but $\eta_C > d_{p,\text{min}}$. Conversely, in the case where a flame was observed (‘FLAME’ in [14]), $\eta_{\text{mix}} > d_p$ and $\eta_C \gg d_p$. In air cases ‘High T_{Air} ’ and ‘Low T_{Air} ’, the mixing length scales are around or bigger than mean particle size $\bar{d}_p$, which leads to the mixing length scale much bigger than the smallest particle size $d_{p,\text{min}}$ used in commercial pulverized coal boilers, $d_{p,\text{min}} \ll \bar{d}_p \lesssim \eta_{\text{mix}}$, consistent with the proposed criterion (8).

3.2. Effects of inlet parameters on the achievement of MILD coal combustion

From the analysis in Section 3.1, Re plays significant roles on scalar mixing around particles. However, the integral length scale plays a role not only in Re but also in the definition of the scalar spectra and therefore in η_{mix} . Figure 3 gives the effects of the inlet velocity (u) and integral length scale (L) on the mixing length scales (η_C and η_B) by solving Eq. (2) and (5) with Eq. (1). Two mixing length scales for $Sc = 0.2$ and $Sc = 1.0$ are compared, shown as red and blue surfaces in the figure, which divide the space into three regimes, ‘MILD’ (above the red surface), ‘Traditional’ (below the blue surface) and ‘Transition’ between them. If particle size is bigger than the smallest mixing length scale for the species with smallest Sc (top red surface), MILD should be achieved. If particle size is smaller than the smallest mixing length scale for the species

with biggest Sc (bottom blue surface), boundary layers will form for all species around the particles and traditional (nonpremixed) combustion is achieved. Between these two cases (red and blue surfaces), a transitional regime is achieved.

Increasing inlet velocity at constant L increases Re and decreases η . On the other hand, increasing L at constant u increases Re and increases η . This is simply a consequence of (1), $\eta = LRe_L^{-3/4} = (\nu^3 L/u^3)^{1/4}$. This explains why relatively small jet diameters and high inlet velocities are used to reach MILD regime in the design of reactor configuration [14,28]. It also provides design guidance. For a specific particle size range, the inlet velocity could be chosen based on the reactor configuration (integral length scale) to reach MILD regime. Alternatively, for specific inlet velocity and reactor configuration (integral length scale), the smallest particle size that could reach MILD regime can be obtained.

We now apply this analysis to literature data. To obtain boundaries for ‘MILD’, ‘Transition’ and ‘Traditional’ regimes for a given particle size, we solve the following two equations to obtain the relationship between u and L for $Sc = 0.2$ and $Sc = 1.0$:

$$d_p = \begin{cases} \eta = LRe_L^{-3/4} = (\nu^3 L/u^3)^{1/4}, & Sc = 1.0 \\ \eta_C = \eta Sc^{-3/4} = LRe_L^{-3/4} Sc^{-3/4}, & Sc = 0.2 \end{cases} \quad (10)$$

We choose d_p to be the smallest reported value in each experimental case and use ν consistent with each experiment as well.

Figure 4 shows the resulting regime diagram for experiments from [11,13,14]. The red and blue lines in Fig. 4 represent MILD boundaries for the smallest particle size in the experiment at $Sc = 0.2$ and $Sc = 1.0$, respectively. This delineates the minimum

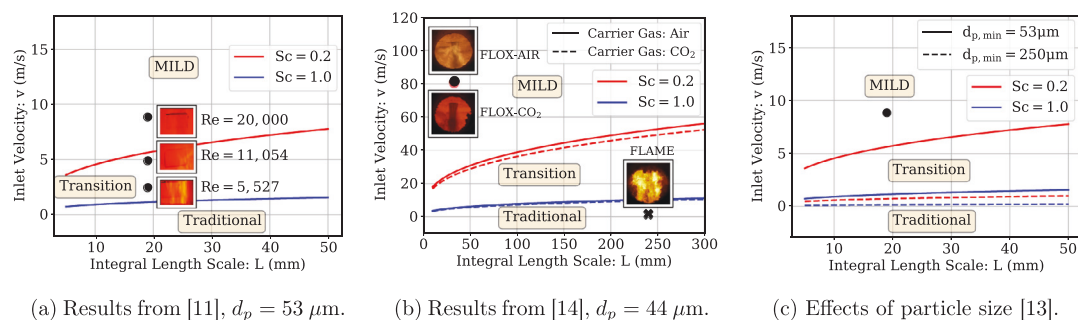


Fig. 4. Verification for influences of inlet parameter using literature data. A fixed particle size is used to calculate the combustion regime boundaries in (a) and (b). Two different particle sizes are used to get solid and dashed regime boundaries in (c). Viscosity, ν , of air and CO_2 as carrier gas are used to get the regime boundaries (solid and dashed lines) in (b). The area above the red solid/dashed line is MILD regime. Data and flame images from [11], [14] and [13] are applied in every sub-figure, respectively. All parameters are set to be the same as the literature expect for inlet velocity, u , and integral length scale, L . The $\bullet$ points indicate the cases where MILD is reported. The $\times$ point indicates the flame case.

inlet velocities for the particles with size d_p to reach MILD regime as a function of the integral length scale. The red and blue lines also divide parameter space into ‘MILD’, ‘Transition’ and ‘Traditional’ regimes.

For example, case $Re_{jet} = 20,000$ in Fig. 4(a) falls into the MILD regime, consistent with experimental observations where no visible flame is observed in the reactor. However, cases with $Re_{jet} = 11,054$ and 5527 in Fig. 4(a) falls into the ‘Transition’ regime, which is reflected by the luminous intensity in the reactor (see also the discussion in Section 3.1 in connection with Fig. 2e–f).

In Fig. 4(b), combustion regimes divided by solid lines apply to ‘FLAME’ and ‘FLOX-AIR’ cases, which use air as the carrier gas, while combustion regimes divided by dashed lines apply to ‘FLOX- CO_2 ’ case, which use CO_2 as the carrier gas. The ‘FLAME’ case falls into the ‘Traditional’ regime with obvious flame shown in the reactor, while the ‘FLOX-AIR’ and ‘FLOX- CO_2 ’ cases are in the ‘MILD’ regime with no visible flame structure shown in the reactor. In Fig. 4(c), small (solid lines) and large (dashed lines) particle sizes are used in Eq. (10). Larger particle size results in a larger ‘MILD’ regime and reduces the transition region. This is consistent with the experimental observations which reported MILD for both particle sizes.

4. Conclusions

In this work, we propose an additional criterion for MILD coal combustion informed by the scalar mixing spectra, and evaluate this against data available in the literature. Our proposed criterion that the smallest mixing length scale (affected by Re and Sc) needs to be of the order of or smaller than the particle size, $\eta_{mix} \lesssim d_p$ (to breakup the heat and mass transfer boundary layers around particles and get well-mixed reactants), is consistent

with literature observations of MILD combustion and can discriminate between cases where MILD and traditional combustion are reported. Furthermore, it provides additional insight into ‘transition’ regime between MILD and traditional combustion based on the range of Sc . Species with $Sc < 1$ together with small particles relative to the Obukhov–Corrsin scale is the main reason for the existence of faint flames in this regime.

The influence of the inlet velocity and jet diameter (integral length scale) are analyzed and verified by the literature data based on our proposed criterion. Higher inlet velocity and smaller jet diameter is helpful to reach MILD regime. This provides general guidance for reactor design based on the particle size and integral length scale or velocity.

Declaration of Competing Interest

None.

Acknowledgments

This research was funded by the National Science Foundation under grant NSF1704141.

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