Analysis of Coal Moderate or Intense Low-Oxygen Dilution (MILD) Combustion

E.-M. Wartha*[1], M. Bösenhofer[1,2], M. Kiss[2], and M. Harasek[1]

[1]Technische Universität Wien, Institute of Chemical, Environmental and Bioscience Engineering, Getreidemarkt 9/166, 1060 Wien

[2]K1-Met GmbH, Area 4 - Simulation and Analysis, Stahlstraße 14, Betriebsgsgebäude (BG) 88, 4020 Linz

Abstract

Moderate or intense low-oxygen dilution offers a great opportunity to reduce harmful pollutants. Besides its wide application for gaseous and liquid combustion, it is also of interest for solid fuels, such as coal. In contrast to gaseous MILD combustion, there is no general agreement on the criteria to (a-priori) analyze and characterize MILD combustion. Therefore, we investigate several suggested criteria based on a well-known coal MILD combustion furnace. The conclusions derived from the different criteria are compared in order to assess their applicability. Additionally, we simulate the furnace with the open-source CFD tool OpenFOAM to calculate the Damköhler number in the overall domain of the furnace.

Introduction

Moderate or intense low-oxygen dilution (MILD) is a promising technology to reduce the emissions in combustion systems and is therefore receiving increasing attention in the research community. Generally speaking, the features of MILD combustion are a low temperature increase, distributed reaction zones and flameless combustion. Therefore, this type of combustion is also called flameless combustion [1]. Cavaliere and Joannon [2] discuss the physical, chemical and thermodynamic aspects of MILD combustion in detail and provide a widely accepted definition of MILD combustion, Eq. (1) and Eq. (2).

Although, most MILD combustion applications can be found for gaseous or liquid fuels, such as industrial furnace applications, gas turbines, internal combustion engines or homogeneous charge compression ignition, MILD combustion can generally be applied to various fuels. Most cases, where solid fuels are burned under MILD conditions are discussed for coal, for example in coal firing plants [3]. The combustion of coal under MILD conditions is also often referred to as high temperature air combustion or flameless oxidation [4]. Although the characterization for MILD combustion by Cavaliere and Joannon [2] is not restricted to gaseous and liquid fuels, the definition is ambiguous for solid fuels [5]. Therefore, several additional criteria have been suggested to characterize solid MILD combustion.

In the following, we compare several of those to characterize coal MILD combustion based on a well-known lab-scale experiment in the IFRF furnace.

Theory

When considering definitions for MILD combustion, the most widely accepted **criteria for MILD combustion** are the one suggested by Cavaliere and Joannon [2]:

$$T_{in} > T_{si} \tag{1}$$

$$\Delta T = T_{WSR} - T_{in} < T_{si} \tag{2}$$

where T_{in} is the inlet temperature, T_{si} the self-ignition temperature and T_{WSR} the temperature in the well-stirred reactor.

Often, the investigation of coal MILD combustion is restricted to the gaseous products of the coal, or even only the volatile components released from the coal [6], because the coal gasification and oxidation are only a surface phenomena and will not feature distributed characteristics.

One ambiguity when considering the criteria by Cavaliere and Joannon [2] for coal combustion is that the inlet temperature T_{in} of the gaseous products from the coal is not clearly defined. In contrast to gaseous combustion, the inlet temperature depends on the heat-up and devolatilization kinetics of the coal. Therefore, Zhou et al. [5] suggest to simulate the heat up of a coal particle in a plug flow reactor (PFR) and consequently estimate the inlet temperature. However, the inlet temperature from a PFR simulation might not coincide with the actual inlet temperature in the application.

Several additional criteria to complement Eq. (1) and Eq. (2) have been suggested to account for the specifics of solid combustion. Zhou et al. [6] proposed a **criterion based on mixing lengths** to address the requirement of mixing the gaseous products of coal combustion and devolatilization, and the surrounding gas/oxidizer. This criterion assesses, if the turbulent mixing scales η_{mix} are in the order of or smaller than the particle size to break up the boundary layer structures around the particle and ensure sufficient mixing. The criterion is defined as follows:

$$\eta_{mix} = \min(\eta_B, \eta_C) \leq d_p, \tag{3}$$

where d_p is the particle diameter, η_B the Batchelor scale and η_C the Obukhov-Corrsin length scale [6]. They are

*Corresponding author: eva-maria.wartha@tuwien.ac.at

calculated based on the Schmidt number Sc and the Kolmogorov length scale η:

$$\eta_B = \eta \mathrm{Sc}^{-1/2} \tag{4}$$

$$\eta_C = \eta Sc^{-3/4}. \tag{5}$$

Similarly, Feng et al. [7] suggest to use an additional **criterion based on mixing times**:

$$\tau_{mix} < c \cdot \tau_i \tag{6}$$

where the mixing time scale τ_{mix} shall be smaller than the product of the turbulent flow characteristics c and the ignition time scale τ_i.

The mixing timescale is in essence the time scale of the integral vortex τ_L, which is the ratio of the turbulent kinetic energy k and the dissipation rate ε:

$$\tau_{mix} = \tau_L = \frac{k}{\varepsilon}. \tag{7}$$

The characteristics of the turbulent flow c are defined as the ratio of the mixing time scale and the Kolmogorov time scale τ_η:

$$c = \frac{\tau_L}{\tau_\eta} = \frac{k/\varepsilon}{(\nu/\varepsilon)^{1/2}} \tag{8}$$

where ν is the kinematic viscosity.

The ignition time scale is defined as the difference between inlet temperature T_i and particle temperature T_p divided by the heating rate and is approximated by the convective heating rate, as suggested in [7].

The criteria presented in [6] and [7] both suggest a similar additional condition by estimating the mixing at the particle range scale in relation to the overall mixing rate of the turbulent flow. However, the criterion by [5] additionally takes into account the Schmidt number and therefore, also accounts for diffusional mixing effects. On the other hand, Feng et al. [7] also consider the heating rate to calculate the particle ignition time for the suggested additional criterion.

Besides those extensions of the classical MILD combustion criteria (Eq. (1) and (2)), we can also characterize the flow by the **Damköhler** number. The Damköhler number relates the mixing τ_{mix} and chemical timescale τ_{chem}:

$$\mathrm{Da} = \frac{\tau_{mix}}{\tau_{chem}}. \tag{9}$$

The combustion regime can be considered MILD, when the Damköhler number is in the range of, or below unity [8]. The chemical time scale is the inverse of the Eigenvalues of the Jacobian Matrix from the ODE-system describing the chemical reactions [9, 10]. However, when there are multiple reactions, the fast and precise approximation of a characteristic chemical time scale is not straightforward, see [11]. To characterize the chemical time scale we use the following time scale approximation:

$$\tau_{chem} = \min_{\dot{\omega}_i<0} \left(\frac{Y_i}{\dot{\omega}_i} \right) \tag{10}$$

where Y_i is the mass fraction of a species and $\dot{\omega}_i$ its production rate. In the IFRF case the time scale is evaluated for the volatile combustion reaction.

Comparing the condition from Eq. (6) we can also rearrange the equation:

$$\frac{\tau_{mix}}{c\tau_i} < 1, \tag{11}$$

which is basically equivalent to the Damköhler number condition, if:

$$\tau_{chem} = c\tau_i. \tag{12}$$

Methods

The previously presented criteria for MILD combustion will be analyzed based on a classical coal combustion case: the IFRF 0.58 MW furnace [4]. This setup represents a classical MILD coal combustion case and has been widely used in literature, e.g. [12–18]. The basic setup of the furnace consists of a round jet, where the preheated air/combustion products and two fuel jets for the coal transported by cooled air enter, see Fig. 1. The boundary conditions for the furnace and the mass fractions of the inflow streams are listed in Table 1. Wall temperatures are set to 1523 K.

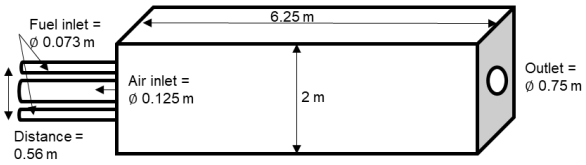

Figure 1: Schematic representation of the IFRF furnace

Table 1: Mass flow rates, inlet temperatures and weight fractions of the inlet streams in the IFRF furnace [14]

		coal	air	ox.	
mass flow	kg/h	66	130	675	
temperature	K	313	313	1623	
		O_2	N_2	CO_2	H_2O	NO
transport air		23	77	-	-	-
oxidizer		22	56	12.5	9.5	89e-4

The used coal is a highly bituminous coal – the Guasare coal – with the proximate and ultimate analysis shown in Table 2.

Table 2: Proximate (FC = fixed carbon, VM = volatile matter, Moist = moisture, HHV = higher heating value in MJ/kg) and ultimate analysis of the Guasare coal [13]

	FC	VM	Ash	Moist	HHV
prox.	56.7	37.1	3.3	2.9	38.307
	C	H	O	N	S
ult.	80.97	5.39	11.26	1.54	0.85

To calculate the volatile species composition, the calibration tool called "pyrolysis kinetic preprocessor (pkp)" [19, 20] was used. The necessary input parameters are the coal properties (Table 2) and representative heating rates for the application: $1.31 \cdot 10^5$, $1.78 \cdot 10^5$ and $2.55 \cdot 10^5$ K/s with maximum temperatures of 1623, 2100 and 2100 K, respectively. The tool is based on the chemical percolation devolatilization (CPD) model [21]. The resulting composition of the volatiles consists of C_2H_4, C_6H_6, H_2O, CO and CO_2. For varying equivalence ratios ($\phi = 0.1, 0.2, 1, 2, 5$) the composition is shown in Fig. 2.

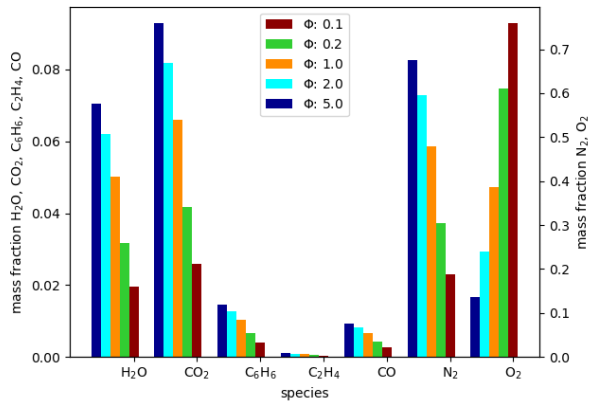

Figure 2: Different concentrations computed by the pkp-tool for the Guasare coal with varying equivalence ratios.

First of all, we analyze the combustion regime based on the criteria Eq. (1) and Eq. (2). For the evaluation, simulations in a well-stirred reactor (WSR) are performed. Here, python using the library cantera [22] and the kinetic mechanism for C1-C3 chemistry [23–25] was used. The volume of the WSR was set the same as the IFRF furnace volume.

The CFD simulation was conducted with the finite volume solver OpenFOAM – version 9 [26]. Due to symmetry reasons only a fourth of the geometry was meshed, as done by [13–15, 17]. The mesh consists of $4.7 \cdot 10^5$ mainly hexahedral cells. For the simulation, the k-ε turbulence model was chosen and the Eddy Dissipation Concept (EDC) accounts for turbulence-chemistry interactions.

The adapted Westbrook-Dryer mechanism [27] models the chemical reactions in the gas phase. Methane was substituted by the pseudo-volatile component with $CH_{3.14}N_{0.064}S_{0.015}O_{0.41}$. The devolatilization was modeled by the competing two-step model. The parameters were fitted by the pkp tool: $A_1 = 90367$, $E_1 = 4.44 \cdot 10^7$, $\alpha_1 = 0.57$, $A_2 = 10^6$, $E_2 = 1.88 \cdot 10^{10}$, $\alpha_2 = 0.34$. The gasification and oxidation of the coal were modeled by a kinetic-diffusion limited approach according to [28].

Results

In the following section the presented criteria:

- Classical criteria [2]: $T_{in} > T_{si}$ & $\Delta T < T_{si}$
- Criterion based on mixing lengths [6]: $\eta_{mix} \leq d_p$
- Criterion based on mixing time [7]: $\tau_{mix} < c \cdot \tau_i$
- Damköhler number analysis: $\mathrm{Da} = \frac{\tau_{mix}}{\tau_{chem}}$

are analyzed based on the coal combustion case in the IFRF furnace.

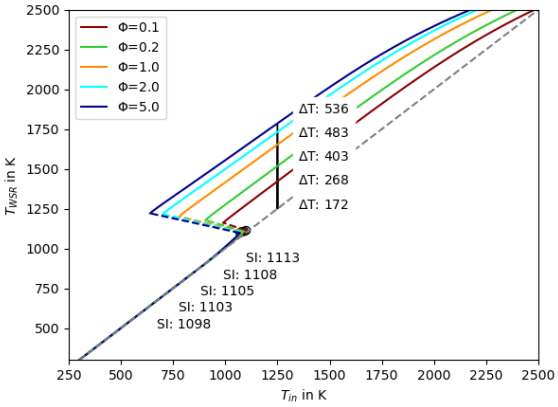

Figure 3: Ignition plot with different equivalence ratios for the Guasare coal

For the composition of volatiles (Fig. 2) the ignition temperatures calculated in the WSR are shown in Fig. 3. The self-ignition temperatures hardly vary with decreasing equivalence ratio (from 1098 K to 1113 K). However, the S-curves become increasingly unfolded for leaner conditions. This means, the leaner the condition, the easier the classical MILD criteria is fulfilled, compare [29]. Here, for all the conditions, we see a conditionally MILD regime, also for an equivalence ratio of 1.4, which corresponds to the IFRF furnace. This suggests, that the inlet temperature should be above 1098 K to achieve MILD conditions.

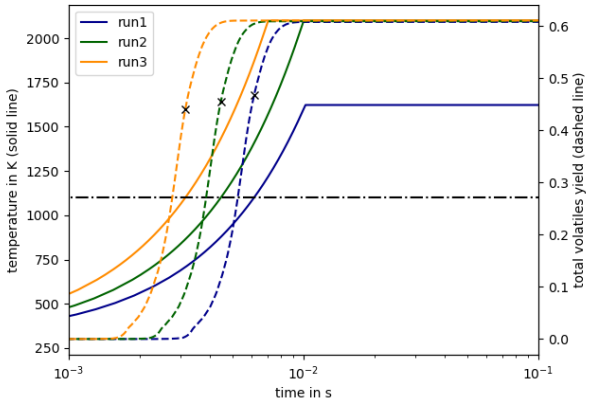

Figure 4: Total volatile yield calculated with the CPD model. The cross marks the volatile yield at self-ignition temperature.

If the fuel inlet temperature is taken as the inlet temperature of the coal, i.e. 313 K, it is clearly too low.

However, Zhou et al. [5] mention that the inlet temperature of the volatiles should be taken as the temperature at which the volatiles are released from the coal.

We could analyze the inlet temperature with the CPD model – using the same heating rate runs as used for the pkp calculations. Fig. 4 shows the temperature profiles and the volatile yields calculated by the CPD model. For those cases, only a small fraction of the volatile release is happening above the self-ignition temperature (marked by cross). Therefore, only for this part the classical MILD criteria would be fulfilled.

Additionally, Fig. 3 shows that ΔT is increasing with increasing equivalence ratio. However, the criterion from Eq. (2) is fulfilled for all the scenarios.

Fig. 5 shows the energy spectra for the IFRF conditions and the particle diameter in correlation to the MILD combustion criterion calculated based on [6]. Following Eq. (3), this additional criterion for coal MILD combustion is fulfilled for the IFRF furnace for all particle diameters and for a range of different Schmidt numbers (shown for 1 and 0.2).

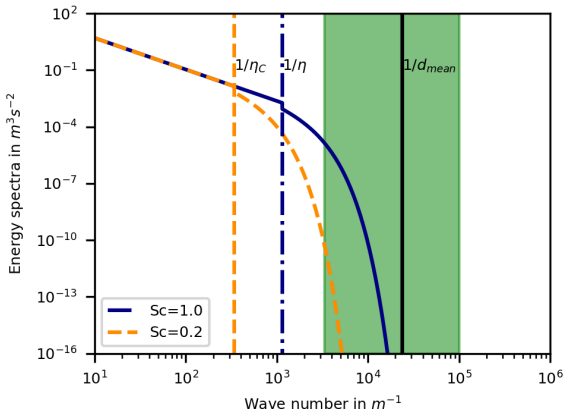

Figure 5: Energy spectra calculated according to [6] with $\nu = 1.8 \cdot 10^{-3}, \varepsilon = 10^4$

Fig. 6 shows the results calculated based on Eq. (6). The criterion proposed by [7] suggest, that MILD combustion only occurs for particle diameters greater than $16\mu m$ – of course, only if the original criteria are also fulfilled. Although, some of the coals are smaller, the majority of the particles – approximately $\geq$ 90 % – of the coal particles are above this threshold. Therefore, this criterion is satisfied.

The CFD simulation was used to analyze the Damköhler number distribution in the furnace. To ensure the correct prediction, Fig. 7 shows the comparison of mole fractions obtained from simulation and experiments [4]. The profiles show a good agreement between simulation and experiment. The CO_2 mole fraction is underpredicted slightly, but overall the trend is well reproduced and, therefore, the simulation results can be used to analyze the Damköhler number distribution.

Fig. 8 shows the Damköhler number distribution calculated from the CFD simulation. The regions with Damköhler numbers above unity are marked in greyscale. Overall of the domain features Damköhler numbers below unity - especially the reactive parts below a height of three meter.

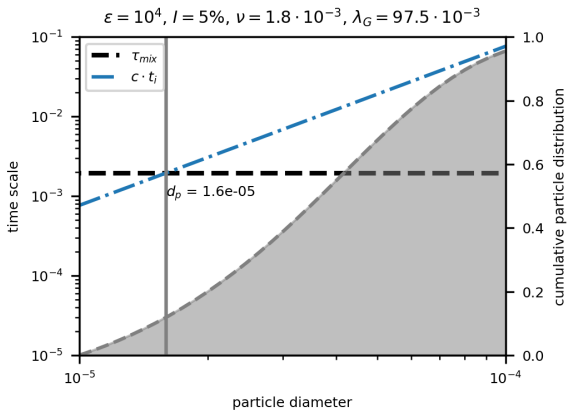

Figure 6: Additional MILD criterion from [7]. The grey area shows the cumulative particle distribution. The criterion is fulfilled for diameters larger than $16\mu m$.

Conclusion

The analysis of the IFRF furnace – which is often taken as a representative coal MILD combustion case – shows, that the classical MILD criterion is only fulfilled for part of the volatiles released from the coal. For part of the released volatiles, the inlet temperature is too low. This contradicts the assumption that this reference case features MILD combustion. However, for part of the volatiles the criteria is fulfilled and some of the model assumptions, such as the devolatilization rate or the heat of devolatilization influence the inlet temperature.

The additional criteria analyzing the turbulent mixing and length scale, both promote the existence of a MILD combustion regime in the furnace. Additionally, the Damköhler number analysis also shows values in the range of or below unity, which characterize MILD combustion regimes.

In conclusion, the classical MILD combustion criteria can help to check the regime a-priori. However, the calculation of the inlet temperatures is not straightforward for coal combustion and requires several a-priori assumptions. The suggested additional criteria can definitely help to ensure well mixing of released gaseous products of the coal and consequently promote MILD combustion. Finally, the Damköhler number analysis is a good way to check the conditions with a CFD-simulation, but it requires more effort compared to the zero-dimensional analyses.

Acknowledgments

The authors gratefully acknowledge the funding support of K1-MET GmbH, metallurgical competence center. The research program of the competence center K1-MET is supported by COMET (Competence Center for Excellent Technologies), the Austrian program for competence centers. COMET is funded by the Federal

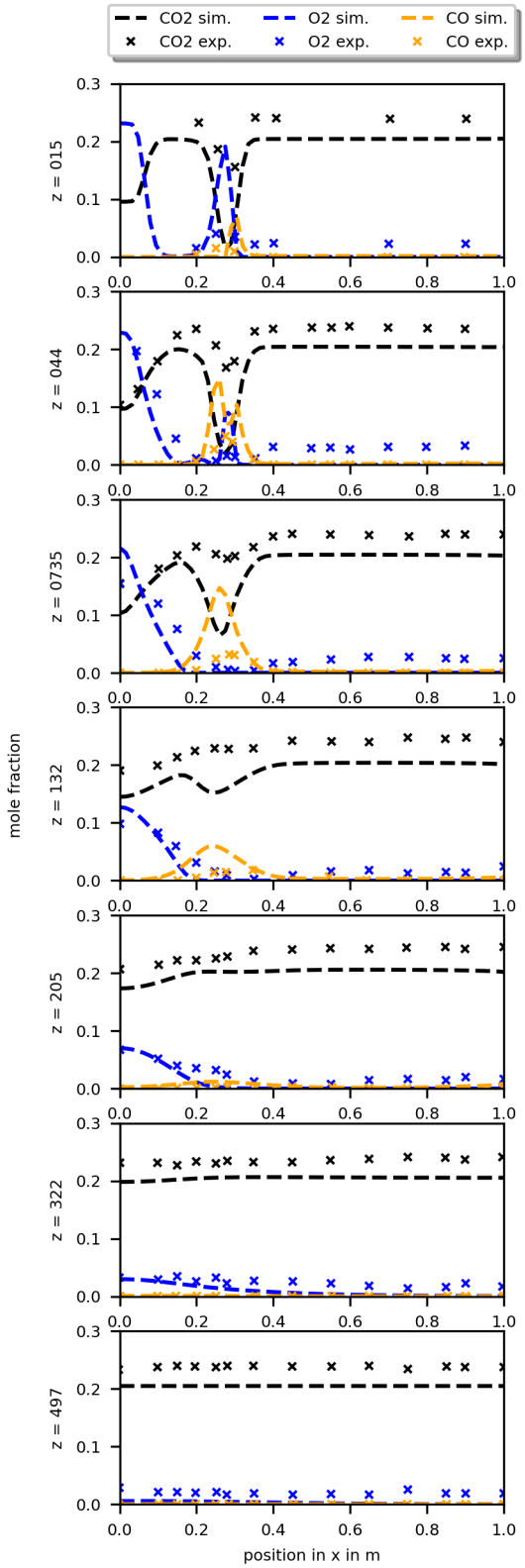

Figure 7: Mole fractions at different positions in the IFRF furnace - comparison of simulation results and experimental results [4]. - Update simulation not converged yet

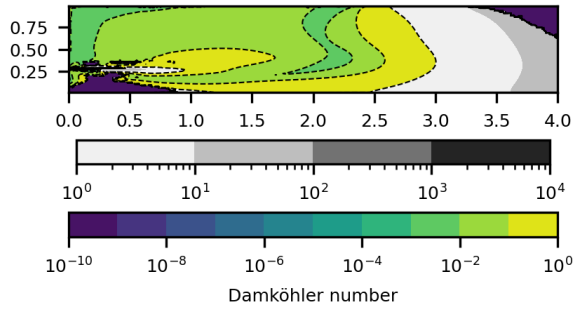

Figure 8: Damköhler number in the IFRF furnace. Values above unity are shown in greyscale.

Ministry for Transport, Innovation and Technology, the Federal Ministry for Science, Research and Economy, the provinces of Upper Austria, Tyrol, and Styria, and the Styrian Business Promotion Agency. The computational results presented have been achieved in part using the Vienna Scientific Cluster (VSC).

References

[1] J. F. Driscoll, J. H. Chen, A. W. Skiba, C. D. Carter, E. R. Hawkes, and H. Wang, Premixed flames subjected to extreme turbulence: Some questions and recent answers (2020).

[2] A. Cavaliere and M. D. Joannon, Mild combustion, *Progress in Energy and Combustion Science* **30**, 329 (2004).

[3] W. P. Adamczyk, R. A. Bialecki, M. Ditaranto, P. Gladysz, N. E. L. Haugen, A. Katelbach-Wozniak, A. Klimanek, S. Sladek, A. Szlek, and G. Wecel, Cfd modeling and thermodynamic analysis of a concept of a mild-oxy combustion large scale pulverized coal boiler, *Energy* **140**, 1305 (2017), ISSN 03605442.

[4] N. Schaffel-Mancini, Ecological evaluation of the pulverized coal combustion in htac technology (2009).

[5] H. Zhou, J. McConnell, T. A. Ring, and J. C. Sutherland, Characterization of temperature criteria using gas-phase fuel streams for mild coal combustion, *Fuel* **296**, 120445 (2021), ISSN 00162361.

[6] H. Zhou, T. A. Ring, and J. C. Sutherland, in *Proceedings of the Combustion Institute* (2021), vol. 38, 4233–4240, ISSN 15407489.

[7] L. Feng, Q. Zhang, Y. Wu, W. Huang, and H. Zhang, Theoretical analysis on the criteria of mild coal combustion, *Energy and Fuels* **33**, 11923 (2019), ISSN 15205029.

[8] M. T. Lewandowski, A. Parente, and J. Pozorski, Generalised eddy dissipation concept for mild combustion regime at low local reynolds and damköhler numbers . part 1 : Model framework development, *Fuel* 117743 (2020), ISSN 0016-2361.

[9] A. S. Tomlin, L. Whitehouse, and M. J. Pilling, Low-dimensional manifolds in tropospheric chemical systems, 125–146 (2002).

[10] T. Nagy and T. Turányi, Relaxation of concentration perturbation in chemical kinetic systems, *React.Kinet.Catal.Lett.* **96**, 269 (2009).

[11] E.-M. Wartha, M. Bösenhofer, and M. Harasek, Characteristic chemical time scales for reactive flow modeling characteristic chemical time scales for reactive flow, *Combustion Science and Technology* **00**, 1 (2020), ISSN 0010-2202.

[12] M. Mancini, R. Weber, and U. Bollettini (2002), vol. 29, 1155–1163.

[13] J. P. Kim, U. Schnell, G. Scheffknecht, and A. C. Benim, Numerical modelling of mild combustion for coal, *Progress in Computational Fluid Dynamics* **7**, 337 (2007), ISSN 14684349.

[14] N. Schaffel, M. Mancini, A. Szlęk, and R. Weber, Mathematical modeling of mild combustion of pulverized coal, *Combustion and Flame* **156**, 1771 (2009), ISSN 00102180.

[15] M. Vascellari and G. Cau, Influence of turbulence-chemical interaction on cfd pulverized coal mild combustion modeling, *Fuel* **101**, 90 (2012), ISSN 00162361.

[16] Z. Mei, P. Li, F. Wang, J. Zhang, and J. Mi, Influences of reactant injection velocities on moderate or intense low-oxygen dilution coal combustion, *Energy and Fuels* **28**, 369 (2014), ISSN 08870624.

[17] Z. Zhang, B. Lu, L. Zhang, X. Li, C. Luo, Y. Xu, Z. Zhao, and C. Zheng, Computational study on the effect of gasification reaction on pulverized coal mild combustion diluted by n2 and co2, *Applied Thermal Engineering* **158** (2019), ISSN 13594311.

[18] Y. Kuang, B. He, W. Tong, C. Wang, and Z. Ying, Effects of oxygen concentration and inlet velocity on pulverized coal mild combustion, *Energy* **198** (2020), ISSN 03605442.

[19] M. Vascellari, R. Arora, M. Pollack, and C. Hasse, Simulation of entrained flow gasification with advanced coal conversion submodels. part 1: Pyrolysis, *Fuel* **113**, 654 (2013), ISSN 00162361.

[20] M. Vascellari, Pyrolysis kinetic preprocessor.

[21] T. H. Fletcher, Review of 30 years of research using the chemical percolation devolatilization model (2019).

[22] D. G. Goodwin, H. K. Moffat, I. Schoegl, R. L. Speth, and B. W. Weber, Cantera: An object-oriented software toolkit for chemical kinetics, thermodynamics, and transport processes, `https://www.cantera.org` (2022), version 2.6.0.

[23] E. Ranzi, A. Frassoldati, A. Stagni, M. Pelucchi, A. Cuoci, and T. Faravelli, Reduced kinetic schemes of complex reaction systems: Fossil and biomass-derived transportation fuels, *International Journal of Chemical Kinetics* **46**, 512 (2014).

[24] E. Ranzi, C. Cavallotti, A. Cuoci, A. Frassoldati, M. Pelucchi, and T. Faravelli, New reaction classes in the kinetic modeling of low temperature oxidation of n-alkanes, *Combustion and Flame* **162**, 1679 (2015), ISSN 0010-2180.

[25] G. Bagheri, E. Ranzi, M. Pelucchi, A. Parente, A. Frassoldati, and T. Faravelli, Comprehensive kinetic study of combustion technologies for low environmental impact: Mild and oxy-fuel combustion of methane, *Combustion and Flame* **212**, 142 (2020), ISSN 0010-2180.

[26] H. Weller, G. Tabor, H. Jasak, C. Fureby, A. You, M. A. Y. Be, and I. In, A tensorial approach to computational continuum mechanics using object-oriented techniques, *Computers in Physics* **12**, 620 (1998).

[27] J. Andersen, C. L. Rasmussen, T. Giselsson, and P. Glarborg, Global combustion mechanisms for use in cfd modeling under oxy-fuel conditions, *Energy and Fuels* **23**, 1379 (2009), ISSN 08870624.

[28] Z. Zhang, B. Lu, Z. Zhao, L. Zhang, Y. Chen, C. Luo, and C. Zheng, Heterogeneous reactions behaviors of pulverized coal mild combustion under different injection conditions, *Fuel* **275** (2020), ISSN 00162361.

[29] M. T. Lewandowski, C. Netzer, D. R. Emberson, and T. Løvås (2021).