Detailed simulations for flamelet modelling of SO$_x$ formation from coal

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This paper reports on recent progress in the modelling of pulsed coal flames. A brief review of current research on fully-resolved and Euler-Lagrange simulations in the context of flamelet modelling of pulsed coal combustion is given first. This is followed by more recent results from the flamelet modelling of SO$_x$ formation due to fuel-S in pulsed coal flames.

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1 Introduction

Despite its well known environmental impact, pulsed coal combustion (PCC) is a major source of primary energy conversion worldwide. As the replacement of coal-fired power stations by renewables and other sources of energy is a long term effort, current research on PCC needs to focus on a) improving efficiency and b) reducing pollutant emission from coal combustion. Due to the hostile environment in PCC chambers reliable, non-intrusive measurements that would help to address these issues are difficult to obtain. Alternatively, numerical simulations can be used to study PCC and to improve our understanding of the physical and chemical phenomena that control the conversion process. This paper reports on recent progress in PCC modelling and is structured as follows. The first part of the paper briefly reviews recent work on PCC modelling, with a focus on fully-resolved and Euler-Lagrange simulations of single coal articles, arrays of particles, and particle clouds in the context of flamelet modelling for large eddy simulation (LES). The second part describes our recent attempts to predict SO$_x$ formation from coals with significant amounts of sulfur by means of flamelet modelling [1].

2 Recent work on PCC modelling

The flow in PCC chambers at the industrial scale is strictly turbulent. Classically, turbulence closures for coal flames have been obtained in the Reynolds-Averaged Navier Stokes (RANS) framework [2]. More recently the potentially higher predictive capability of LES approaches for turbulence modelling has been investigated, where mostly flamelet approaches have been employed to model subgrid scale turbulence-chemistry interactions [3–5]. However, flamelet-LES modelling of PCC flames is not straightforward, since the mixing of several fuel (volatiles and char off-gases) and oxidiser streams needs to be accurately described [6] and because strong enthalpy changes due to gas-solid heat transfer must be considered [7]. To aid the development of flamelet models for PCC detailed simulations that attempt to reach the limit of direct numerical simulation (DNS) can be used. DNS aims to resolve all relevant physics of the problem and is meant to be model-free. While it is accepted that DNS can be performed for single phase combustion, full DNS of coal flames seems difficult to achieve. This is due to the added complexity in PCC, where -among other factors- the role of porous media, devolatilisation and char conversion, homogeneous and heterogeneous kinetics, as well as population balance equation (PBE) effects like particle breakage, agglomeration, growth/shrinkage etc. need to be considered. As including these effects requires further (complex) sub-modelling, here the terms fully-resolved and Euler-Lagrange simulations are preferred over DNS. Fully-resolved simulations resolve all scales of turbulence, the flame and the boundary layers around individual particles, whereas Euler-Lagrange simulations neglect the boundary layers and revert to the tracking of Lagrangian point particles. In the following sections 2.1 and 2.2 recent fully-resolved and Euler-Lagrange simulations are summarised and references to flamelet modelling are made. Due to space constraints this review is limited to recent 2D and 3D simulations of reacting flows with coal and does not include related work from 1D and non-burning approaches.

2.1 Fully-resolved simulations

Recently a number of fully-resolved simulations of single particles [8–11] and coal particle arrays [12–14] have been performed. Vascevali et al. [8] pioneered the work on fully-resolved simulations of single coal particle ignition experiments [15] for flamelet modelling. The authors found a good agreement with the experimental ignition delay time and were able to model the fully-resolved data with a flamelet approach, if the fully-resolved scalar dissipation rate profiles were employed. Farazi et al. [10] simulated single char particle combustion studying the effects of particle size, relative velocity and gas atmosphere on particle burning. This work was later extended to char particle arrays, where the impact of inter-particle distance and

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particle arrangement was analysed [12]. Tufano et al. [9] conducted fully-resolved simulations of single coal particle ignition experiments [15] in various gas atmospheres. The work confirmed the earlier findings in [8] and demonstrated that homogeneous particle ignition always occurs in the downstream wake region behind the particle regardless of gas composition. The experimental trends of ignition delay times in various N\textsubscript{2}/O\textsubscript{2}/CO\textsubscript{2} mixtures were captured and the sensitivity of absolute ignition delay time predictions to gas phase chemistry, particle Reynolds number \(R_{ep}\), particle preheat and extraction criteria for ignition delay were highlighted. Vascellari et al. [11] modelled the fully-resolved data of [9] with a flamelet approach, in both \textit{a priori} and \textit{a posteriori} simulations. The flamelet table was based on unsteady laminar diffusion flamelets and a novel theoretical formulation of the scalar dissipation rate in the vicinity of single particles allowed for accurate flamelet predictions of the ignition process. Tufano et al. [13, 14] performed fully-resolved simulations of coal particle arrays in both laminar and turbulent flows. The sensitivity of the simulation results to transient effects and varying inter-particle distance (or global equivalence ratios) was studied in [13]. A wide range of burning conditions was observed, spanning from individual particle burning (for large spacing/lean mixtures) to group combustion (for small spacing/rich mixtures). The effects of increasing particle \(R_{ep}\) and turbulent flow were investigated in [14]. It was found that elevated \(R_{ep}\) leads to the formation of wake flames due to extinction events in the highly strained upstream region and that conventional flamelets do not capture the flame structure at large \(R_{ep}\) correctly. Turbulence was found to enhance the range of chemical states near the particles and led to a local increase of chemical turnover for the most upstream particle layer, but reduced chemical conversion for downstream particles due flame flapping and localised extinction. In summary, a number of recent fully-resolved simulations of volatile matter and char combustion provide detailed reference data for PCC-LES modelling. While the fully-resolved data on flame ignition, flame structure and particle interactions is vast, little information on pollutant precursor formation is yet available.

### 2.2 Euler-Lagrange simulations

Fully-resolved simulation approaches provide the most detailed description of the direct vicinity of burning particles, but they are restricted to small computational domains and can only consider a limited part of the turbulent spectrum. Alternatively, the particle boundary layers may be neglected and Euler-Lagrange simulations or \textit{carrier-phase} DNS (CP-DNS) can encompass larger turbulent spectrum, as well as relative motion between particles. Coal particle burning in laminar flow has been studied by Euler-Lagrange simulations in [16–21]. In a series of papers Wen et al. [16–19] studied laminar counterflow combustion of coal relevant for flamelet modelling. The main conclusions were that (i) predictions are sensitive to a number of model and physical parameters (e.g. volatile matter composition, homogeneous chemistry, particle size, particle loading, fluid strain, radiative heat transfer), (ii) coal flames are significantly different from pure (non-premixed) gas flames, with the former potentially exhibiting double flame structures with both premixed and non-premixed combustion regimes and (iii) flamelet tabulation methods for PCC should - at least - consider the governing variables total mixture fraction (volatile + char off-gases), a mixing parameter, progress variable and normalised enthalpy. Knappstein et al. [20] studied volatile combustion from single particles in laminar flow and compared reference direct chemistry simulations to a premixed flamelet (FGM) approach. They found sensitivities to the assumed volatile composition and small deviations of their FGM result from the reference near the particle only. Farazi et al. [21] studied coal particle stream ignition in laminar flow using a detailed devolatisation model and varied coal loading, gas temperature and velocity. It was found that low gas temperature, high coal loading and large slip velocity delay ignition and that velocities can change significantly due to the mutual interaction of the two phases. Hara et al. [22] performed CP-DNS of a coal particle jet flame using tailor-made homogeneous two-step chemistry. They saw a reasonable agreement of the DNS with experiments for both velocities and scalars and found an inner premixed reaction zone converting volatiles and O\textsubscript{2}, and an outer non-premixed combustion zone where volatiles react with O\textsubscript{2} and CO. Similarly, Bai et al. [23] performed CP-DNS of a coal jet flame and confirmed the occurrence of mixed-regime combustion, where strong reactions are located in regions of low vorticity and high scalar dissipation rate. They emphasised the additional effect of devolatisation on scalar dissipation and identified three typical coal jet flame structures referred to as interspersed flame, stripe flame and stable continuous flame. Brosh et al. [24] performed CP-DNS of localised forced ignition of coal particle-laden mixtures for various particle equivalence ratios, turbulence levels and particle diameters. They identified both premixed and non-premixed combustion zones and found that overly rich mixtures - due to high particle equivalence ratios or fast devolatisation from small particles - promote extinction and that mild turbulence promotes combustion, whereas strong turbulence suppresses it. Rieth et al. [25] conducted CP-DNS of coal combustion in a reacting shear layer of coal particles entrained in air (upper stream) mixing with hot lean combustion products (lower stream), employing a relatively detailed volatile composition and homogeneous chemistry. Ignition was found in lean regions around particles entrained into the lower oxidising stream and at late times combustion occurred in the non-premixed mode in a lower flame, whereas partially premixed combustion with significant two-phase heat transfer was observed at the edge of particle-laden upper stream. A subsequent study addressed steady non-premixed flamelet/progress variable (FPV) modelling of the same CP-DNS dataset, both \textit{a priori} and \textit{a posteriori} [26]. It was found that the lower non-premixed flame can be captured well by steady non-premixed FPV, whereas FPV predictions of the upper flame deviate more strongly from the direct chemistry solution due to unsteady and premixing effects. Finally, Wan et al. [27] performed CP-DNS of a 3D planar turbulent jet with sodium-containing coal particles using both direct chemistry and FGM tabulation. They found ignition to mainly occur in partially premixed zones, whereas combustion primarily proceeds in diffusion-like reactive layers. Sodium species predictions from direct chemistry...
and FGM tabulation agreed generally well. In summary, a number of PCC Euler–Lagrange simulations has been published over the past four years. The simulations focused largely on ignition, volatile combustion and flame structure analysis, and evaluations of flamelet approaches versus direct chemistry solutions have been performed. However, with the exception of [27] relatively little focus has yet been set on the prediction of pollutant precursor formation.

3 Flamelet modelling of SO\textsubscript{x} formation

Pulverised coal power plants are considered as a major source of SO\textsubscript{x} (including SO, SO\textsubscript{2} and SO\textsubscript{3}) emission since coal may contain significant amounts of chemically bound sulfur, yet not many detailed (i.e. fully-resolved or Euler–Lagrange) studies of SO\textsubscript{x} formation resulting from fuel-S are available. The purpose of this work twofold. First, the recently developed detailed SO\textsubscript{x} formation mechanism by Gersen et al. [28] (including 63 sulfur-containing species) and the reduced mechanism by Cerru et al. [29] (including 12 sulfur-containing species) [29] are compared for combustion in a plug flow reactor (PFR). For better comparability to our earlier work the SO\textsubscript{x} formation mechanisms are embedded in a core C–H–O oxidation mechanism for homogeneous chemistry [9, 25]. Following this, our earlier PCC–FPV model [16] is extended to consider SO\textsubscript{x} formation and results from direct chemistry (DC) integration and FPV tabulation are compared within Euler–Lagrange simulations of PCC in a laminar counterflow burner.

3.1 Modelling approach

We compare results from detailed chemistry simulations that directly solve for the transport of mass, momentum, total enthalpy and all species mass fractions to an FPV approach that only solves for the governing variables of the tabulated chemical manifold. The mixture fractions for volatiles, Z\textsubscript{vol}, and char off-gases, Z\textsubscript{pro}, are defined as Z\textsubscript{vol} = ξ\textsubscript{vol} / (ξ\textsubscript{vol} + ξ\textsubscript{pro} + ξ\textsubscript{ox}) and Z\textsubscript{pro} = ξ\textsubscript{pro} / (ξ\textsubscript{vol} + ξ\textsubscript{pro} + ξ\textsubscript{ox}), where ξ\textsubscript{vol}, ξ\textsubscript{pro} and ξ\textsubscript{ox} are the mass of gas originating from the volatiles, char off-gases and oxidiser stream. To avoid possible numerical issues, the coordinates Z\textsubscript{vol} and Z\textsubscript{pro} are transformed to the new coordinates X and Z defined as X = Z\textsubscript{vol} / (Z\textsubscript{vol} + Z\textsubscript{pro}) and Z = Z\textsubscript{vol} + Z\textsubscript{pro}. In addition to X and Z the flamelet library contains two more trajectory variables, namely the normalized total enthalpy H\textsubscript{norm} and the reaction progress variable Y\textsubscript{PV}. Finally, the flamelet solutions Ψ are parameterized as Ψ = Ψ (X, Z, H\textsubscript{norm}, Y\textsubscript{PV}). Methods for the flamelet modelling of SO\textsubscript{x} formation in PCC can be divided into two principal categories, a) extracting sulfur species from the flamelet library directly and b) solving transport equations for sulfur species mass fractions, in which only the reaction source terms are extracted from the table. The governing equation for the SO\textsubscript{x} mass fractions reads

\[
\frac{∂ (ρ ϕ_i \text{SO}_x)}{∂ t} + \frac{∂ (ρ u_j ϕ_i \text{SO}_x)}{∂ x_j} = \frac{∂}{∂ x_j} \left( ρ D_{ϕ_i \text{SO}_x} \frac{∂ ϕ_i \text{SO}_x}{∂ x_j} \right) + ω_{i \text{SO}_x}, \tag{1}
\]

where ϕ\textsubscript{i \text{SO}_x} represents the SO\textsubscript{x} species mass fractions, including Y\textsubscript{SO}, Y\textsubscript{SO\textsubscript{2}} and Y\textsubscript{SO\textsubscript{3}}. ω\textsubscript{i \text{SO}_x} is the SO\textsubscript{x} reaction rate that can be extracted from the table using the formation/consumption rates ω\textsubscript{i \text{SO}_x} and ω\textsubscript{i \text{SO}_x}, cf. Tab. 1, as described in [30].

| Table 1: Employed FPV tabulation methods for SO\textsubscript{x} modelling |
|-----------------------------|-----------------------------|
| a) M1 Extracting Y\textsubscript{SO}, from the flamelet table directly, ϕ\textsubscript{1 \text{SO}_x} = Ψ (X, Z, H\textsubscript{norm}, Y\textsubscript{PV}) |
| a) M2 Extracting Y\textsubscript{SO\textsubscript{2}}, from the flamelet table directly, ϕ\textsubscript{2 \text{SO}_x} = Ψ (X, Z, H\textsubscript{norm}, Y\textsubscript{PV}) |
| b) M3 Solving Eq. (1), ω\textsubscript{1 \text{SO}_x} = Ψ (X, Z, H\textsubscript{norm}, Y\textsubscript{PV}) |
| b) M4 Solving Eq. (1), ω\textsubscript{2 \text{SO}_x} = Ψ (X, Z, H\textsubscript{norm}, Y\textsubscript{PV}) |

Considering that the formation process from SO\textsubscript{2} to SO\textsubscript{3} is slow, the conventional reaction progress variable Y\textsubscript{PV}\textsubscript{conv} that only consists of the major reaction products H\textsubscript{2}O and CO\textsubscript{2} is reformulated as Y\textsubscript{PV}\textsubscript{modi} = Y\textsubscript{H\textsubscript{2}O} + Y\textsubscript{CO\textsubscript{2}} + λY\textsubscript{SO\textsubscript{3}}, with scaling parameter λ = 1000. The flamelet solutions can be extracted either using Y\textsubscript{PV}\textsubscript{conv} or Y\textsubscript{PV}\textsubscript{modi}. Combing the methods in categories (a) and (b), four flamelet tabulation methods are evaluated in this work, as summarised in Tab. 1.

3.2 Results and discussion

The SO\textsubscript{x} reaction mechanisms developed by Gersen et al. [28], referred to as “DTU”, and by Cerru et al. [29], referred to as “STU” are evaluated in a PFR operated at T\textsubscript{0} = 1400 K and φ = 2 first. The comparison between these two mechanisms is shown in Fig. 1a. It is found that for the intermediate species CO the predictions from the two mechanisms agree well, while for SO\textsubscript{3} slight differences can be observed. The overall agreement between the two mechanisms (not shown) indicates that the SO\textsubscript{x} formation process can be characterised to sufficient accuracy by the simplified STU mechanism, which is subsequently used in the DC counterflow simulation and to generate the flamelet library. Subsequently, the flamelet models are evaluated for a 2D counterflow PCC flame, the computational set-up of which can be found in [16] (case 4 therein). The FPV results are compared to the detailed chemistry solutions along the central axis of the counterflow burner. Figure 1b compares the SO\textsubscript{3} mass fraction between different flamelet tabulation methods and the DC solution. It is found that extracting SO\textsubscript{3} from...
the flamelet library with the conventional and modified reaction progress variables (M1 and M2) shows negligible difference. Furthermore, it is seen that solving the transport equation for \( \dot{Y}_{SO_3} \) does not improve the prediction accuracy, regardless of the method (M3 and M4) used to obtain the reaction source term. This indicates that the steady flamelet model can be used to predict \( \dot{SO}_3 \) formation in pulsed coal flames, despite the slow conversion process from \( SO_2 \) to \( SO_3 \).

4 Summary and conclusions

A brief review of recent developments in fully-resolved and Euler-Lagrange simulations for PCC has been given. Two \( SO_2 \) reaction mechanisms and different FPV tabulation methods for \( SO_3 \) formation from PCC have been compared, establishing the smaller mechanism as sufficient and motivating the need for further research into detailed FPV modelling of \( SO_3 \) formation.

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