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# Conditional moment closure modelling of soot formation in turbulent, non-premixed methane and propane flames

Robert M. Woolley *, Michael Fairweather, Yunardi

*Energy and Resources Research Institute, School of Process, Environmental and Materials Engineering, University of Leeds, Leeds LS2 9JT, UK*

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ABSTRACT

Presented are results obtained from the incorporation of a semi-empirical soot model into a first-order conditional moment closure (CMC) approach to modelling turbulent, non-premixed methane–air and propane–air flames. Soot formation is determined via the solution of two transport equations for soot mass fraction and particle number density, with acetylene and benzene employed as the incipient species responsible for soot nucleation, and the concentrations of these calculated using a detailed gas-phase kinetic scheme involving 70 species. The study focuses on the influence of differential diffusion of soot particles on soot volume fraction predictions. The results of calculations are compared with experimental data for atmospheric and 3 atm methane flames, and propane flames with air preheated to 323 K and 773 K. Overall, the study demonstrates that the model, when used in conjunction with a representation of differential diffusion effects, is capable of accurately predicting soot formation in the turbulent non-premixed flames considered.



## 1. Introduction

Despite dwindling resources, fossil fuel combustion still plays a major role in the world economy and is widely used for the production of energy. The formation and emission of particulate pollutants such as soot, as a consequence of hydrocarbon combustion, is fast becoming a major concern in both developed and more so, developing countries. Soot generation usually results from incomplete combustion and typically occurs at fuel-rich stoichiometries. Although some of these particles are oxidized in the flame, soot that escapes oxidation is considered a serious environmental pollutant. There are also associated health risks since both polycyclic aromatic hydrocarbons (PAHs) that are precursors of soot and soot-associated organics have been identified to be carcinogenic. On the other hand, in cases where soot oxidation is completed within a flame, higher production of intermediate soot is desirable for increasing the radiant heat transfer from flames. Thus, control of soot production and reduction of soot emission from combustion processes are problems that need to be solved to obtain effective heat generation and to reduce harmful impacts to humans and the environment. Quantitative understanding of the soot growth and oxidation mechanisms in flames are critical to the development of approaches to control soot emissions.

Most practical combustion systems such as the gas turbine and internal combustion engine operate at high turbulence levels, with or without a combination of high pressure and preheated air. Consequently, it is important to numerically investigate soot formation under these conditions. While operation at elevated pressures proffers the significant advantage of increasing the thermodynamic efficiency of the system, it is also disadvantageous due to the releasing of more soot particles into the environment. The pressure dependence of soot formation and oxidation mechanisms is complicated, and there is some evidence that they may be dissimilar for different pressure levels and hydrocarbon fuels. Although they are still not fully understood, it is widely accepted among researchers that increasing the pressure in the environment surrounding a non-premixed flame alters the reaction rate and the diffusion coefficients, which lead to the increase of soot production [1]. Most experimental studies of sooting processes to date have focussed primarily on laminar flames at atmospheric pressure and thus available data on soot levels in turbulent non-premixed flames at elevated pressures is very limited. Measurements in laminar [2,3] and turbulent [4] non-premixed flames have shown that soot formation increases and soot oxidation decreases with increasing pressure. Brookes and Moss [4] argued that the increase of soot production from flames of 1 to 3 atm in pressure was due to the increase of the density and species concentrations (acetylene in particular), in the elevated-pressure conditions, as well as the increase of soot residence time. In contrast to operation at high pressure which produces more soot, air preheating has been used as a means of reducing soot emissions and optimizing fuel consumption in practical combustors. As the air temperature becomes higher, the rates of some elementary reaction steps increase, while

* Corresponding author. Tel.: +44 (0) 113 343 2351; fax: +44 (0) 113 343 2405.
*E-mail address:* r.m.woolley@leeds.ac.uk (R.M. Woolley).

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**Nomenclature**

| | |
|---|---|
| $d$ | diameter |
| $k$ | reaction rate constant or turbulence kinetic energy |
| $p$ | partial pressure |
| $r$ | radial distance or reaction rate |
| $u$ | axial velocity |
| $x$ | axial distance along centre-line |
| $A$ | surface area or pre-exponential factor |
| $C$ | constant |
| $D$ | diffusion coefficient |
| $M$ | molar mass |
| $N$ | number or particle number density |
| $P$ | probability density function |
| $Q$ | transported scalar |
| $R$ | radius |
| $T$ | temperature |
| $Y$ | mass fraction |

*Greek symbols*

| | |
|---|---|
| $\varepsilon$ | dissipation rate of k |
| $\eta$ | sample space variable |
| $\kappa$ | Boltzmann constant |
| $\nu$ | kinematic viscosity |
| $\xi$ | mixture fraction |
| $\rho$ | density |
| $\sigma$ | Stefan–Boltzmann constant |
| $\tau$ | characteristic time scale |
| $\varphi$ | integration variable |
| $\chi$ | scalar dissipation |
| $\omega$ | production rate |

*Subscripts*

| | |
|---|---|
| fv | soot volume fraction |
| g | growth |
| $h$ | enthalpy |
| $i$ | reactive scalar index |
| n | nucleation |
| ox | oxidation |
| $s$ | soot |
| A | Avogadro number |
| K | Kolmogorov viscous scale |
| $R$ | radius |

*Superscripts*

| | |
|---|---|
| + | scalar of equal diffusivity |
| $*$ | cross-stream averaged |

*Other symbols*

| | |
|---|---|
| $\langle\rangle$ | ensemble averaging |
| $\{\}_R$ | integration over cross section limited by $\|r\| < R$ |
| $\langle\alpha\|\beta\rangle$ | conditional expectation of $\alpha$ at some value $\beta$ |
| $\sim$ | Favre average |

those of other elementary steps decrease. These changes affect the distribution of species and temperature across the flame as well as soot, CO, and NO emissions. A matter of concern in high-temperature air combustion is that of $NO_x$ emissions. Experimental studies in to non-premixed methane flames [5,6] have shown that as the temperature of incoming combustion air is increased, the $NO_x$ emissions also increase. A major obstacle for further development of a combustion system with high efficiency and low soot emissions therefore appears to be the trade-off between the effects of elevated pressure and increased air preheat.

One of major challenges in turbulent combustion modelling involves accounting for interactions between turbulent flow and complex, finite-rate chemistry that are of profound importance when the prediction of minor chemical species and pollutants such as $NO_x$ and soot is desirable. Significant progress has been made within the last two decades toward the development of combustion models capable of representing such interactions in turbulent combustion applications. Amongst those proposed, two appear to offer the most promising features for future development; these being the transported probability density function (PDF) approach [7] and the conditional moment closure (CMC) method [8]. Both achieve the goal of the inclusion of finite-rate chemistry into turbulent flow calculations, the former model doing so via the solution of a multi-dimensional transport equation of species and vector quantities, with solutions obtained by the implementation of a Monte–Carlo technique. Although displaying very promising results in similar applications [9,10], the method is computationally expensive, especially when the number of chemical species is large. At present, the deterministic CMC method provides a less restrictive approach to a wide range of practical applications, and can be easily integrated with a complex chemical kinetic scheme without requiring significant computer run times. Roomina and Bilger [11] investigated the application of CMC in modelling an attached turbulent methane flame and reported good agreement with data, with the exception to NO. Similar discrepancies in NO prediction were observed by Fairweather and Woolley [12,13], who used a first-order CMC model to predict attached turbulent non-premixed flames of methane and hydrogen. Further investigation [14] revealed that by applying a second-order CMC closure to the chemical source term, improvement of NO predictions could be obtained. Calculations of lifted flames [15,16] have also been made, with reasonable agreement obtained with experimental data. Kim et al. [17] compared the performance of the CMC approach with that of the stationary laminar flamelet model when predicting the more complex flow of a bluff-body stabilized methanol flame, finding that CMC results provide superior agreement with data. In addition to these successes in modelling gas-phase combustion, CMC has previously shown promise in the calculation of soot formation in non-premixed flames [18].

In this paper, the results of an application of a first-order CMC approach [8] to the calculation of turbulent non-premixed flames and soot formation are presented. The soot model used in the calculations is based on that presented by Leung et al. [19] and Lindstedt [20], with transport equations for soot mass fraction and particle number density incorporated into the CMC approach. The influence of differential diffusion of soot particles in the context of CMC modelling, previously investigated by Kronenburg et al. [18], is further assessed within the computation of methane elevated pressure and propane preheated air flames. The turbulent flow field and CMC results in terms of mixture fraction, temperature, and soot volume fraction or soot concentration are validated against available experimental data [4,21].

## 2. Mathematical modelling

### 2.1. Experimentally investigated flames

The non-premixed elevated-pressure methane, and preheated-air propane, flames considered in the present study were experimentally reported, respectively, by Brookes and Moss [4] and Nishida and Mukohara [21]. The methane–air flames [4] were studied at pressures of 1 and 3 atm. The flame was confined within a cylin-