

SIMULATION AND MODELLING OF REACTIVE PARTICLES IN TURBULENCE

Nigel S. A. SMITH

Aeronautical & Maritime Research Laboratory
506 Lorimer St, Fishermens Bend, Victoria, AUSTRALIA

August 1998

Introduction

Condensed-phase particles are frequently present in turbulent combustion systems and can have a profound influence on the thermochemical nature of their surroundings. Fuel droplets and soot particles are two examples of important condensed phase species in combustion. The evaporation of the former largely determines the distribution of the combustible gaseous fuel/air mixture, while the presence of the latter impacts strongly on the degree of radiant heat transfer from the system. It is thus desirable to be able to predict the evolution of these particles, in a turbulent combusting environment, in response to their local thermochemical conditions.

A number of difficulties surround the modelling of the mean rate of particle phase and chemical reactions in turbulent combustion. Perhaps the most significant difficulty is associated with determining the mean influence of carrier fluid properties such as temperature and chemical species concentrations upon the particle population. Wherever the local properties of the carrier fluid fluctuate according to turbulent mixing action, these fluctuations couple with the non-linearity of the particle reactions to prevent a simple first order closure of the mean reaction rates using mean properties. This type of closure problem is the same as that experienced in modelling the mean rate of chemical reactions in purely gas phase combustion when using conventional averaging techniques.

The Conditional Moment Closure (CMC) method (see Klimenko 1990, Bilger 1993) for modelling turbulent (gas phase) nonpremixed combustion makes use of averages which are conditional upon the local value of a conserved scalar (mixture fraction), which is indicative of the state of mixing between fuel and air masses. Conditional averaging on mixture fraction captures much of the turbulence-induced fluctuations and a first order closure is often possible. The success of the CMC model in gas phase combustion makes it

of some interest in modelling particle reactions.

The purpose of this study was to simulate the dynamics of reactive particle mass and motion in a turbulent combusting environment and attempt to model the observed mean thermodynamic behaviour of the reacting particles using a derivative of the CMC method. In this report, the simulation and modelling of pseudo-soot particles is described. A similar study conducted for pseudo-droplet particles is described by Smith (1998).

Simulation Conditions

The direct numerical simulations were conducted using a pseudo-spectral solution technique for forced isotropic turbulent flow on a 32-cubed grid. The flow was incompressible in nature, but a passive scalar field was used in conjunction with an equilibrium temperature profile in passive scalar space, to determine a false temperature map throughout the domain for the purpose of determining instantaneous particle reaction rates. The passive scalar field was forced at large scales by the interaction of the turbulent motions with an imposed mean scalar gradient in one direction. Cubic spline tensor-products were employed to determine local fluid velocity and temperature at all particle locations. Particle properties were advanced in time using a fourth-order Adams-Bashforth timestepping routine.

The following Lagrangian equation for particle motion was solved,

$$\frac{dv_i}{dt} = \frac{\alpha^*}{\tau_k} (m^*)^{-2/3} (u_i(\underline{x}) - v_i) \quad (1)$$

where v_i is the particle velocity, $u_i(\underline{x})$ is the local fluid velocity at the particle location, m^* is the nondimensional particle mass, τ_k is the Kolmogorov timescale, and α^* is the nondimensional characteristic particle rate given by,

$$\alpha^* = 18 \left(\frac{\rho_f}{\rho_p} \right) \left(\frac{L_k}{d} \right)^2 \quad (2)$$

In the above, L_k is the Kolmogorov length scale, d is the reference particle diameter, ρ_p is the particle material density, and ρ_f is the fluid density.

The rate of change of any given particle's mass was given by

$$\frac{dm^*}{dt} = \frac{\alpha^*}{\tau_k} \beta (m^*)^{1/3} \left(f_{gr} - f_{co} (m^*)^{1/3} \right), \quad (3)$$

where β is a generic rate coefficient, and f_{gr} and f_{co} are normalised functions of local mixture fraction (and thus an inferred reactive gas phase composition) which mimic surface growth and consumption respectively. The growth and consumption reactions were designed to embody the basic features of the soot processes they represent, but with a notable difference. Namely, that all reaction rates were symmetric in mixture fraction space about the mean mixture fraction. Particle surface growth was strongest at the mean mixture fraction, and particle consumption was strongest somewhat to the lean and rich sides of the mean mixture fraction. The exact form of the reaction rates is not at issue in this study.

The reference particle rate (α^*) differed between simulations to reflect different sizes and reactivity of soot particles. The details of the simulation cases studied are given in Table 1.

case	s1	s2
(α^*, β)	(0.7, 40)	(2.8, 1)

Table 1: Simulated characteristic particle rates

In each case, 8192 particles were maintained in the domain, at a constant mean number density, by continuous injection once the forcing had produced a statistically stationary flow field. The simulations were conducted at a Taylor Reynolds number of ~ 24 , and were allowed to proceed over a duration of many eddy turnover times. Statistically stationary particle data were postprocessed from each simulation for comparison with model predictions.

Modelling Method

Application of the CMC model for gas phase turbulent combustion to modelling condensed phase fields requires some consideration of the differences in behaviour between the two phases. Firstly, it was assumed that the particles were, in the mean, small enough to follow the smallest scales of motion. This was found to be largely true in the soot particle simulations, but was not well justified in fuel droplet simulations reported elsewhere (Smith 1998).

Secondly, it is clear that the particle phase does not diffuse appreciably on a molecular level, and thus a the particle phase and mixture fraction are transported in a completely different manner at this level.

The effect of this differential diffusion should be incorporated into the CMC model.

Further, at the smallest scales, a distinct difference between the distribution of gaseous species and condensed phase species is expected (see Klimenko 1990). Significant local structure can exist in the particle field at scales where gas phase fields have been completely smeared by molecular diffusion. The existence of such local structure implies that instantaneous local deviations of particle mass fractions from means, conditional upon gas phase mixture fraction, can be large.

In the present preliminary study, particle effects upon the gas phase are neglected, and the effect of gas phase fluctuations on particle evolution is examined in isolation. However, in reality, the two-way interactions between gas and condensed phase species can be very important. In these instances, large conditional variance in particle properties could lead to similarly elevated levels of conditional variance in gas phase properties and thereby increase the difficulty of the chemical closure problem in the gas phase. It is for this reason that it is important to be able to predict the level of conditional variance of particle mass fraction. The CMC model employed in this study made use of a differential diffusion variant of the second-order conditional moment closure proposed by Li and Bilger (1996). Further details on its derivation can be found in Smith (1998).

Two conditional moment equations were solved in an Eulerian frame in order to represent the reacting particle system. The statistically stationary form of the conditional mean particle mass fraction ($Q_\eta \equiv \langle Y | \eta \rangle$) profile is given by,

$$0 = cN_\eta \frac{\partial^2 Q_\eta}{\partial \eta^2} + (c-1) M_\eta \frac{\partial Q_\eta}{\partial \eta} - \frac{1}{P_\eta} \frac{\partial^2}{\partial \eta^2} (P_\eta C_\eta) + \langle \dot{\omega} | \eta \rangle + I_Q, \quad (4)$$

where N_η is the conditional mean scalar dissipation rate, M_η is the conditional mean scalar diffusion rate, C_η is the conditional covariance of mass fraction and scalar dissipation rate, P_η is the mixture fraction probability density function (PDF), $\langle \dot{\omega} | \eta \rangle$ is the conditional mean reaction rate, I_Q is the conditional mean injection rate, and c is a *mixing mode* variable.

The conditional variance of particle mass fraction ($q_\eta^2 \equiv \langle y^2 | \eta \rangle = \langle Y_\eta^2 | \eta \rangle - Q_\eta^2$) was solved using,

$$0 = cN_\eta \frac{\partial^2 q_\eta^2}{\partial \eta^2} + (c-1) M_\eta \frac{\partial q_\eta^2}{\partial \eta} + 2 \langle y\dot{\omega} | \eta \rangle + S_q + I_q, \quad (5)$$

where I_q is the conditional variance source due to particle injection, and S_q is a grouping of variance

production terms given by,

$$S_q \equiv 2cC_\eta \frac{\partial^2 Q_\eta}{\partial \eta^2} + \frac{2}{P_\eta} (c-1) \frac{\partial}{\partial \eta} (C_\eta P_\eta) \frac{\partial Q_\eta}{\partial \eta} \quad (6)$$

Chemical source terms in Eqns 4 & 5 were determined using instantaneous local temperature (taken from an invariant chemical equilibrium profile), and the first two conditional moments of particle mass fraction at each location in mixture fraction space.

Unclosed terms containing the *slip velocity* between the particle and gaseous fields have been omitted from the equations given above. These terms would otherwise appear as $\left\langle \frac{\partial}{\partial x_i} ((u_i - v_i) y) \mid \eta \right\rangle$ and $\left\langle \frac{\partial}{\partial x_i} ((u_i - v_i) y^2) \mid \eta \right\rangle$ on the right hand side of Eqns 4 & 5 respectively. These unclosed terms increase in magnitude with increasing particle size, and can act so as to obviate the effectiveness of a conditional moment closure for large particles (see Smith 1998).

The *mixing mode* variable, included in the CMC equations above, provides a means of accounting for differential diffusion effects. In the limit where mixing is overwhelmingly due to turbulent stirring, the mixing mode variable tends to unity. In cases where a substantial proportion of the mixing is due to molecular mass transfer, the mixing mode variable will vary substantially from unity. It has been proposed that this variable might be modelled as a function of some global mixing parameter, such as the Kolmogorov scalar scale, independently of mixture fraction and species mass fractions (see Smith 1998). The independence of c of mixture fraction and species mass fractions is borne out by the single value which has sufficed in the studies conducted to date for common mixing conditions but with widely varying particle characteristics.

Kronenburg and Bilger (1997) successfully account for differential diffusion in the CMC model through the use of an alternate treatment from that proposed above. Their methodology has much to recommend it, but when applied to multi-component reacting systems requires an additional conditional moment equation to be solved for each species. The method presented above seeks to avoid this added cost.

The conditional particle injection source terms (I_Q & I_q) are given by,

$$I_Q = \frac{P_I}{\tau_p} (1 - Q_\eta) \quad (7)$$

$$I_q = \frac{P_I}{\tau_p} ((1 - Q_\eta)^2 - q_\eta^2) \quad (8)$$

where P_I is the PDF of particle injection mixture fraction, and τ_p is the time required for a number of injections equal to the size of the particle population to occur (determined from simulation). In the above,

it is assumed that particles are injected to replace those which leave the system, and all freshly injected particles have unit mass.

Results

The characteristic effect on the conditional mean particle mass fraction of varying the mixing mode variable, c , can be seen in Fig. 1 for case *s1*, along with the profile which results from equilibrium between chemical reactions and particle injection alone. It can be seen that the arbitrary increase of c causes the predicted mean mass fraction profile to simultaneously increase in peak value and decrease in minimum value. It can be seen that the equilibrium profile, which is the result of no mixing at all, exhibits the widest range in maxima and minima.

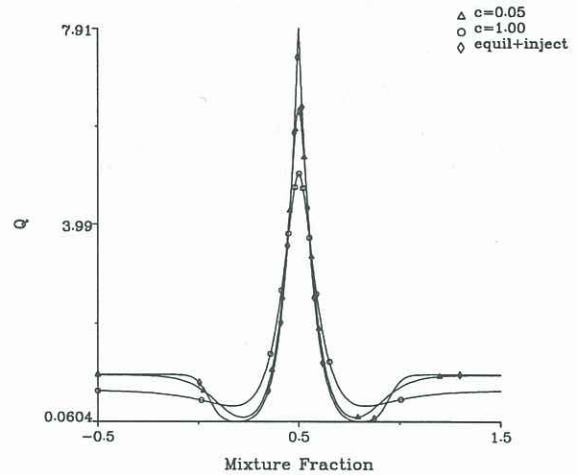


Figure 1: Predicted conditional mean mass fraction profiles for case *s1* with arbitrarily varied levels of differential diffusion, compared with chemical/injection equilibrium profile.

Consideration of the complete set of DNS data generated, suggests that a mixing mode variable value of approximately $c = 0.05$ (minimum attainable) provides the best agreement for all model predictions under the mixing conditions studied.

Predicted and observed conditional mean particle mass fractions are compared in Fig. 2 for cases *s1* and *s2*, with the above mentioned mixing mode. It can be seen that despite the sizeable difference in particle size and reactivity between the two simulation cases, the CMC model provides good overall profile agreement.

In modelling the conditional mean and variance, the model closure of Li and Bilger (1996) for the conditional covariance of scalar dissipation rate and reactive scalar (C_η) was found to be inappropriate. The simulation data revealed the characteristic form of the covariance profile (see Fig. 3) as possessing a large negative spike near the mean mixture fraction. An alternative model for C_η was devised which largely captured this behaviour (see Smith 1998). The mod-

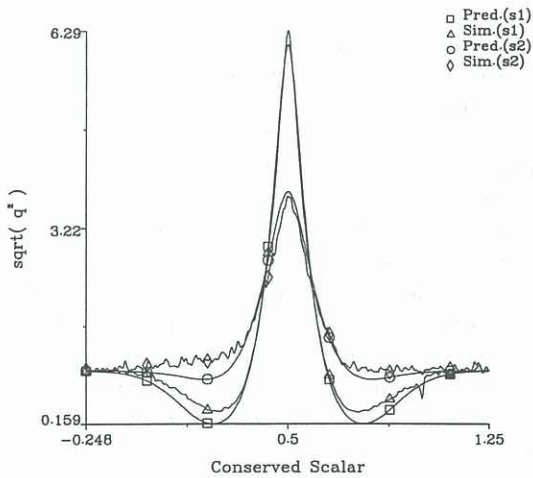


Figure 2: Predicted and observed conditional mean mass fraction profiles for cases $s1$ and $s2$ with $c = 0.05$

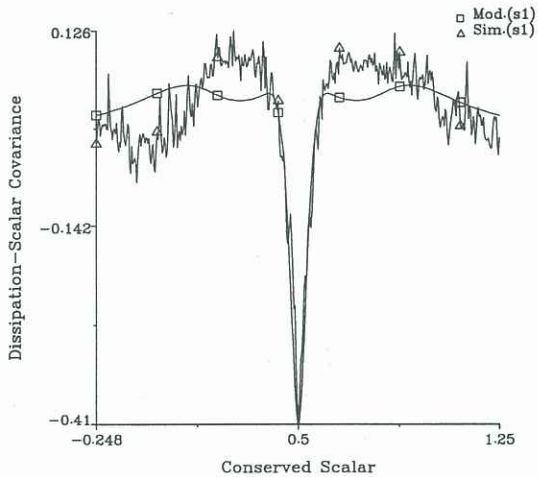


Figure 3: Modelled and observed conditional covariance between mass fraction and scalar dissipation rate for case $s1$.

elled conditional covariance profile is also plotted in Fig. 3, where it can be seen that match between the profiles is effective around the mean mixture fraction, but is quite poor elsewhere.

A comparison of predicted and observed conditional root mean square deviation ($\sqrt{q_\eta^2}$) of mass fraction, can be drawn from Fig. 4. It is evident that there is good qualitative agreement between the predicted and observed profiles, with the CMC model capturing the essential change in profile form between the simulated cases. Good quantitative agreement is found near the mean mixture fraction in both cases, however the predicted profiles deviate substantially from the observed data at mixture fractions away from the mean. This discrepancy was probably due to the poor modelling of the conditional covariance term (C_η) at the same mixture fractions.

Remarks

The results of this preliminary study indicate that a

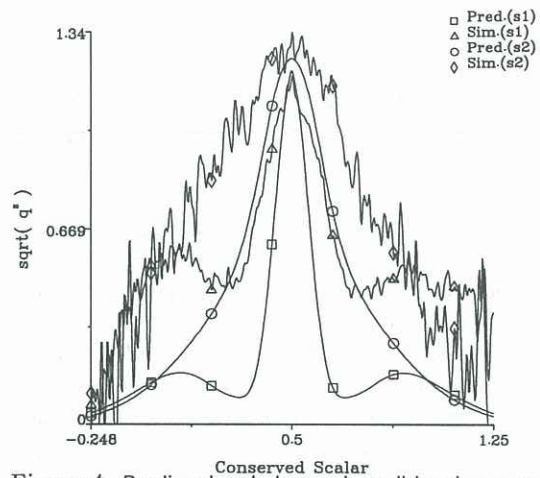


Figure 4: Predicted and observed conditional root mean square mass fraction profiles for cases $s1$ and $s2$ with $c = 0.05$

tailored variant of the Conditional Moment Closure (CMC) method can be applied successfully to modelling the evolution of soot-like reacting particles in a turbulent combusting environment. This finding indicates that it is appropriate to simultaneously model soot and gas phase reactions in turbulence using the CMC model within an Eulerian frame.

A large amount of further work is warranted in this area. The improved prediction of conditional reactive particle behaviour requires a superior means of modelling the conditional covariance (C_η). Further, the mixing mode variable approach to treating differential diffusion in the CMC model requires further attention. A model for the mixing mode variable, as a function of global mixing parameters, is being tested by studying simulation data for similar particle types under parametrically varied turbulent mixing conditions.

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