REDUCED CHEMICAL MODELS FOR INCLUSION INTO CALCULATION METHODS FOR TURBULENT REACTING FLOWS

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ABSTRACT

The coupling of chemistry with computational fluid dynamic simulations of turbulent reacting flows will for the foreseeable future require the use of scalar space reduction techniques preferably coupled with computationally efficient storage and retrieval routines. The current contribution assesses the practical requirements on systematically reduced chemistry in the context of transported Joint Probability Density Function (JPDF) methods. The latter approach is effectively combustion regime independent and has been shown capable of reproducing features, such as extinction and re-ignition, that are of increasing importance in high performance combustion devices operating closer to stability limits. The stochastic Monte-Carlo based methods used to solve the JPDF typically explore the full available state space and the approach thus provides a good indicator of the overall mathematical tractability of different mechanisms. The results obtained show that for natural gas oxidation good functionality may be retained with around 15 independent scalars resulting in efficiency gains in the range 5 to 10 times compared to the full detailed mechanism. It is argued that efficient storage and retrieval algorithms will be required for significant further gains. The current contribution also explores potential scalar space requirements for multi-component propulsion fuels and it appears likely that, in addition to carefully reduced reaction mechanisms and efficient tabulation techniques, advances in distributed computing will be of key future importance.

BACKGROUND

The application of chemical kinetics to the design and analysis of combustion devices has traditionally been limited to features that may be treated as perturbations on solutions obtained with infinite rate chemistry. Such perturbation approaches are strongly application dependent and generally more complete scalar space descriptions are required. However, the introduction of detailed chemical reaction mechanisms into complex multi-dimensional design computations is not possible at present. The physics of scalar reduction techniques is inevitably implicitly based on the introduction of QSSA approximations (e.g. Turanyi *et al.* 1993) through time-scale analysis. The classical systematic reduction method for spatially homogeneous systems was extended to inhomogeneous systems by Peters (1985) through the introduction of a formal reactive-diffusive operator. A large number of studies have subsequently been performed using this approach. The other current principal techniques are based on the construction of explicit mechanisms through Computational Singular Perturbation (CSP) methods (e.g. Goussis and Lam 1991) or the determination of Intrinsic Low-Dimensional Manifolds (ILDM) (e.g. Maas and Pope 1992). The number of degrees of freedom required in a manifold or CSP-derived mechanism will clearly be similar to the number of species retained in

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explicitly reduced reaction mechanisms. Potentially efficient storage and retrieval techniques include In Situ Tabulation (ISAT) (Pope 1997) and the Piecewise Reusable Solution Mapping (PRISM) method (Tonse *et al.* 1999). Efficient integration techniques are inevitably based on Newton expansions and thus require knowledge of the Jacobian matrix which has a tendency to change rapidly even in "close" proximity to the solution. The use of regime dependent adaptive chemistry techniques (e.g. Schwer and Green 2001) is interesting in this context and is also receiving increasing attention. The extent to which simplifications are possible in turbulent reacting flows is closely linked to the accuracy required in the resolution of turbulence chemistry interactions. The low and high Damköhler number limits may readily be dealt with through established techniques and the current contribution considers the situation where turbulence and chemistry time scales are similar. The ability to reproduce key phenomena becomes much more critical in the latter case and the practical usefulness of reduced mechanisms is also strongly linked to the mathematical properties of the terms used in the numerical integration of the chemical source term. The case of natural gas oxidation, using a mechanism comprehensively validated for all principal fuel components (CH_4, C_2H_6, C_2H_4 and C_2H_2), is considered in some detail and the work also projects forward towards practical multi-component propulsion fuels.

ILLUSTRATIVE RESULTS

Difficulties with systematic reduction techniques arise for heavily reduced mechanisms where steady-state and/or partial equilibrium approximations become questionable on physical grounds. Examples related to the formation of C_2 hydrocarbons under natural gas combustion and the associated NO reburn chemistry (e.g. $HCCO + NO = (HNCO)_{isomers} + CO$) are shown in Figure 1. For the QSSD approximation to apply $(C_k/D_k - 1) \sim 0$ and it is evident that neither ketene nor the HNCO isomers can strictly be regarded as being in steady state at temperatures below 1500 K. It is also evident that the same applies to all C_2 hydrocarbons as well as the methyl radical. The example given may be regarded as typical and illustrates the practical difficulties in balancing accuracy and computational tractability. The results for two laminar partially premixed flames (Barlow *et al.* 2001) are shown in Figure 2. Computations with GRI 3.0 (Bowman *et al.* 1997) and the mechanism of Dean *et al.* (1999) are compared with the starting mechanism (Sick *et al.* 1998) in order to illustrate intrinsic uncertainties. The performance of the systematically reduced reaction mechanism, featuring 16 independent scalars and based on the latter mechanism, is also shown and the agreement is arguably satisfactory. The application of the same mechanism to the equivalent turbulent jet diffusion flame is shown in Figure 3 and the results are very encouraging and support the conclusions of earlier studies (e.g. Lindstedt *et al.* 2000 and Barlow *et al.* 2001). The generalization of such results to more complex fuel mixtures is an interesting topic and a preliminary analysis of a detailed chemical mechanism featuring 339 species and 1966 reaction aimed at multi-component Diesel fuels consisting of mixtures of dodecane, m-xylene, naphthalene, toluene, styrene, n-butyl benzene, indene and 1-methyl naphthalene suggests that around 50 species are in steady state at 1200 K and that a further 140 may be removed without unacceptable loss of accuracy. The resulting computational efficiency gains can again be expected to approach a factor of 10 and further economies can be made on the basis of the elimination of individual fuel components. Simulations of elliptic multi-dimensional time-dependent flows using JPDF techniques with 20 scalars are now possible on modest Linux Clusters (Lindstedt and Kuan 2003). However, to carry around 100 species in practical CFD simulations is not possible at this time and the

derivation of "custom built" simplified mechanism with more limited applicability is likely to be required along with efficient tabulation techniques for economical simulations of practical geometries.

CONCLUSIONS

The efficiency gains that can be expected through scalar space reduction techniques alone are likely to be in the range 5 to 10 for a fully retained functionality. This result is governed by the underlying physics and chemistry and it is therefore unlikely that significantly greater economies can be made without loss of accuracy. Furthermore, the tools used in the derivation of reduced mechanisms (e.g. time-scale analysis) have advanced significantly over the last decade. The principal difficulties are arguably therefore to decide what constitutes acceptable approximations and to ensure mathematical tractability of the (non-) linear algebraic equations for species exhibiting short time-scales. A sensible approach is therefore to build targeted mechanisms for key phenomena of interest (e.g. ignition) that are applicable to a given parameter space. It is additionally suggested that efficient storage and retrieval algorithms will play a key role in further efficiency gains and that it is likely that high performance distributed computing techniques will come to the fore in practical design calculations.

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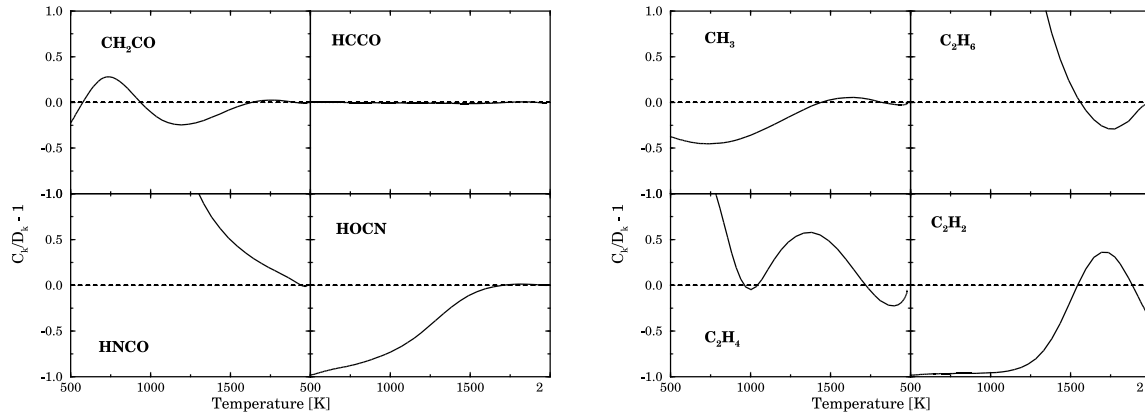
Figure 1. Steady state operators of chemical species related to the reburn of oxides of nitrogen under fuel rich conditions. The simulations correspond to a premixed methane-air flame for φ= 1.4. The graphs have been plotted against flame temperature.
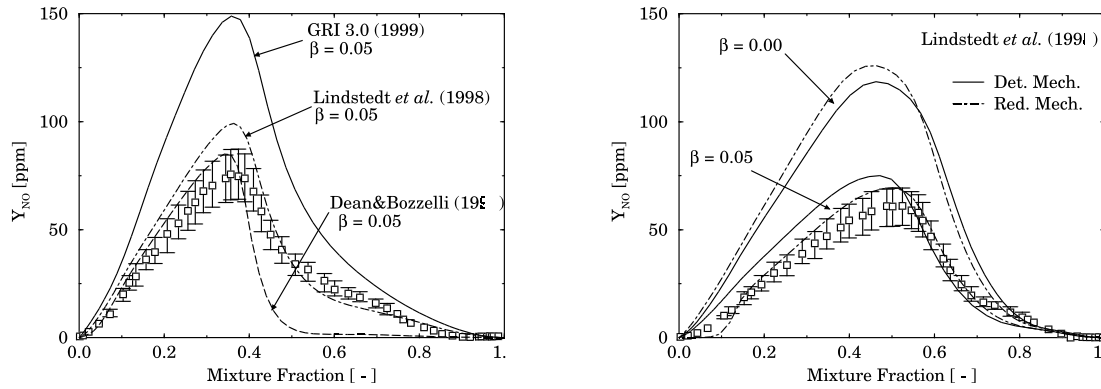

Figure 2. The application of detailed and systematically reduced chemistry to the partially premixed flames reported by Barlow *et al.* (2001). The graph to left compares the uncertainties associated with the NO formation chemistry through the application of three detailed mechanisms. The heat loss factor β is set to 5% in all cases which leads to acceptable agreement with measured temperature profiles. The flame to the right corresponds to φ = 3.17 and that on the right to φ = 2.2.
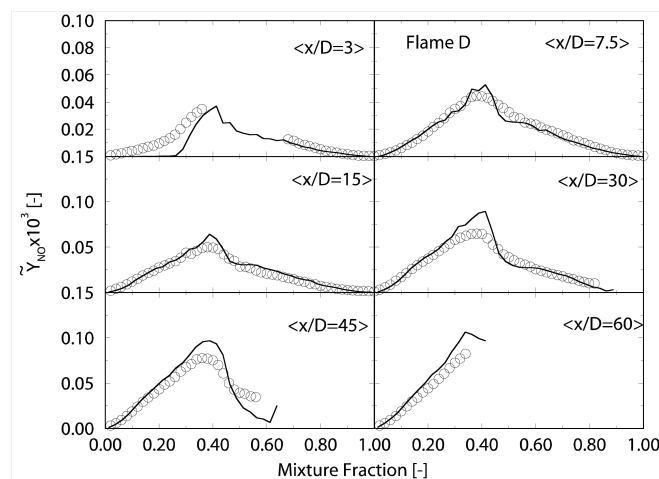

Figure 3. Computations of NO mass fractions for Flame D (Frank *et al.* 2000) by the application of the reduced mechanism corresponding to Figure 2. The computed radiant fraction using RADCAL of 7.4% is in reasonable agreement with the 5.1% estimated by the experimental investigators.