A computational facility for reacting flow science

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Abstract. We discuss recent developments in the application of high-order adaptive mesh refinement constructions in reacting flow computations. We present results pertaining to the time integration of coupled diffusive-convective terms in this context using a stabilized explicit Runge-Kutta-Chebyshev scheme. We also discuss chemical model reduction strategies, with a focus on the utilization of computational singular perturbation theory for generation of simplified chemical models. Starting from a detailed chemical mechanism for methane-air combustion, we examine *a posteriori* errors in flame species computed with a range of simplified mechanisms corresponding to a varying degree of model reduction.

1. Introduction

Computational studies of chemically reacting flows present a number of challenges pertaining to the multiscale nature of the problem and the significant complexity of detailed chemical models of hydrocarbon fuels. The range of length and time scales that are relevant in these flows is large, even when considering relatively simple gas-phase laboratory-scale flames. Moreover, the complexity of the chemical models required for accurate modeling of even the simplest hydrocarbon fuel kinetics is overwhelming. This complexity, coupled with temporal stiffness and the large ranges of length scales, results in a significant computational load in a multidimensional reacting flow solver.

Regarding the range of length-scales in particular, it is important to note that the structure of reacting flows typically involves well-defined thin reaction zones where most chemical processes are active. The structure of these reaction zones in atmospheric hydrocarbon flames typically involves radical profiles with thicknesses on the order of 100μm. Since these fine structures typically exist only in a small region of the computational domain, one may employ adaptive mesh refinement (AMR) [1] to concentrate resolution only where needed [2–4]. Further, many fundamental aspects of reacting flow physics are amenable to study without recourse to complex flow geometries. In such a case, the use of block-structured adaptive meshes [4] is an attractive option. Moreoever, imposing the requirement that the meshes be structured allows efficient data storage and simpler (Cartesian) implementations of various finite-difference stencils.

Block-structured AMR, coupled with high-order spatial discretizations, can serve as a powerful tool to reduce the computational expense of reacting flow computations. High-order methods can deliver the same accuracy as second-order ones (the conventional choice for

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most block-structured AMR constructions) at significantly lower resolutions and consequently, computational cost [5]. This reduction in resolution requirements also assists in reducing the time spent in time-advancing the expensive chemical source terms. In [6], we address many practical issues in using high-order methods in AMR, including the "correct" pairing of interpolations with derivative stencils (as opposed to simply using interpolations of the same order as the derivative stencils) as well as filtering approaches for controlling Runge phenomena introduced by high-order prolongations at coarse-fine interfaces. However, high-order stencils are larger and more cumbersome to use than compact second-order ones; their use, particularly at domain boundaries where they have to be skewed, is particularly challenging. The software developed for the study in [6] and used in [5] has been made available as a library and can be freely downloaded at [7].

Time integration in AMR reacting flow computations poses its own challenges. In particular, implicit time-integration schemes are typically used to integrate diffusive terms in reacting flow equations on block-structured adaptive meshes [4], since diffusional stiffness increases as $(1/\Delta x)^2$ with refinement. On the other hand, extended-stability explicit time integration schemes, e.g. the Runge-Kutta-Chebyshev (RKC) scheme [8], have also been used for the same purpose in flame simulations [9]. However, the original RKC construction is not suitable for time integration of convective source terms. Our investigations into using a Strang-split approach to time-advancing convection and diffusion separately have shown that such constructions result in large temporal errors near domain boundaries when used with time-dependent boundary conditions. Since coarse-fine mesh boundaries exhibit a similar behavior, convection and diffusion are, therefore, best kept coupled.

Beside the utilization of high-order AMR on shallow mesh hierarchies, suitable simplification of detailed chemical models is a key means of improving efficiency of reacting flow computations. There has been much activity in this area in the combustion literature over a number of decades. Two notable reviews present a comprehensive survey of this literature [10, 11]. We have relied on computational singular perturbation (CSP) analysis [12] both for analysis and reduction of chemical systems. CSP enables automatic identification of fast and slow reaction processes, the elimination of fast-exhausted and dormant modes, and the identification of the slow modes that drive the slow evolution of the chemical system. We use CSP for generation of simplifed/reduced chemical models which are then used in subsequent computations as skeletal elementary rate mechanisms [13]. The resulting skeletal mechanisms are not stiffness free, but they do involve a smaller number of reactions and species, relative to the original one, accurately representing both its fast and slow components. This construction employs well-defined error thresholds, thereby controlling accuracy levels in the prediction of a kernel set of species. On the other hand, it is worthwhile inspecting *a posteriori* errors in computed flame solutions with the resulting skeletal mechanisms to provide a more complete understanding of the predictive power of the resulting reduced/simplified models.

In the following, we explore the use of a modified RKC construction for time integration of the coupled transport (convection and diffusion) terms in a high-order AMR context. We also outline the results of a study focusing on the *a posteriori* errors using CSP-simplified chemical models in flame computations.

2. Adaptive Mesh Refinement

As mentioned in Sec. 1, RKC schemes may be used to address purely diffusional stiffness in flame computations. A recent modification [14] to RKC allows moderate amounts of convection to be accomodated, even though the problem is diffusion-dominated. This is achieved by broadening the stability region along the imaginary axis while narrowing it along the real one. Our previous experience with RKC in flame simulations, within an operator-split construction, showed that the timestep restriction is largely set by splitting errors [9]; thus, with flexible choice of the
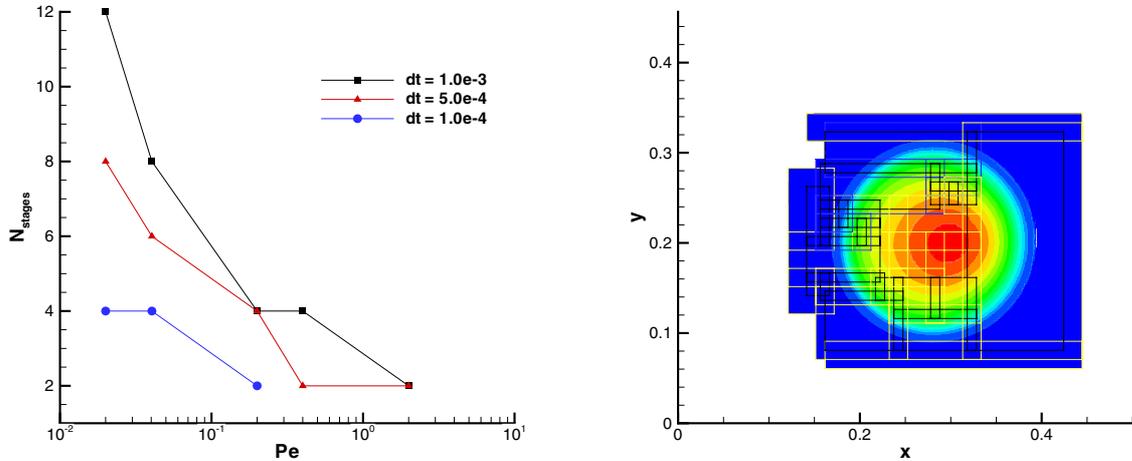
Figure 1. Left: Number for stages versus grid Peclet number for various values of the timestep Δt. The number of stages were obtained from a stability argument. Right: We plot the OH distribution around one of the "hot-spots" in a convective-diffusive-reactive simulation with a constant horizontal advective wind from the right. Only solutions on the 25μm (yellow borders) and 12.5μm (black borders) patches are plotted.

number of internal integration stages, unless the accommodation of the convective term places excessive restrictions on the timestep size, the modified algorithm has the potential for use in flame simulations on block-structured adaptive meshes. In this regard, we solved the viscous Burgers equation on a unit domain at a variety of grid Peclet number ($U\Delta x/\nu$) obtained by varying ν with $U = 2$. In Fig. 1 (left) we plot the optimal number of RKC stages (obtained using the approach in [14]) for a given Δt. We see that the number of stages required for a given Δt reduces as the effect of diffusion decreases; in convection dominated flows, the optimal number of stages is 2 (proved analytically in [14]). Further, as ν increases (and grid Peclet number decreases), we see diffusional stiffness manifest itself and increasing number of stages are required to maintain stability. Also, as Δt decreases, the number of stages required for stability drops. In hydrocarbon flame simulations (with temperatures around 2000 K) the grid Peclet numbers for various species vary between 0.01 and 0.1 on a 10μm mesh, thus indicating a requirement of approximately 10 stages. This is not an excessive increase over typical 8-stage RKC [9] implementations.

We tested the modified RKC algorithm, along with the components of the CFRFS Toolkit [5], to solve a convective-diffusive-reactive problem with hydrogen-air chemistry [15]. "Hot-spots" were initialized in a unit square (1 cm × 1 cm) with a stoichiometric hydrogen-air mixture. A fourth-order spatial discretization approach was used on a four-level block-structured adaptive mesh; the finest grid had a resolution of 12.5 μm. A constant horizontal advective wind from the right was imposed on the domain. In Fig. 1 (right), we plot the OH mass fraction field around one of the hot-spots at time $t = 0.1$ msec. We see an asymmetric profile that exhibits the expected distortion by the advective wind. We are currently exploring the parametric space of this construction and embedding it with a projection solver for the momentum equations.

3. Chemical Model Reduction
We used the CSP-based chemical model reduction/simplification strategy, outlined in [13], to develop a range of chemical mechanisms, with different degrees of reduction, for methane-air flames at atmospheric pressure and room-temperature reactants. This was done starting from

the GRImech3.0 [16] detailed mechanism, using a range of values of the CSP error-tolerance. We evaluated the simplified mechanisms against the starting mechanisms in 1D flame computations, in order to evaluate *a posteriori* errors in flame solutions with different degrees of model simplification. It is important to note that, by construction, the mechanism simplification strategy is designed to guarantee accurate prediction of a set of *kernel* species. In the present study, using the kernel species $\{CH_4, CO_2\}$, we specifically examined errors in other species retained in the mechanisms.
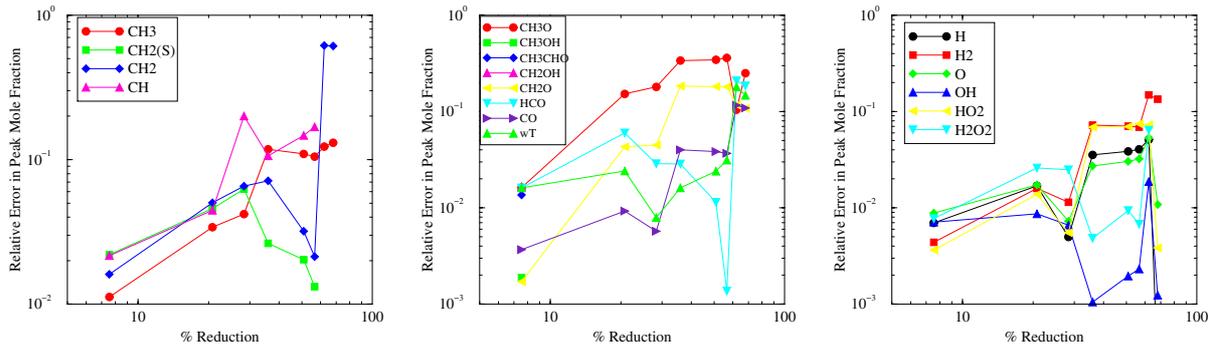

Figure 2. Plots of relative error in peak mole fraction of various species, and heat release rate, in computed 1D premixed methane-air flame solutions based on a range of simplified mechanisms derived from GRImech3.0 [16].

Results corresponding to stoichiometric reactants are shown in Figure 2. The plots in the figure show the observed relative error in various species peak mole fractions, and the heat release rate, in the flame, evaluated relative to GRImech3.0 [16], for different degrees of model reduction. The degree of model reduction is defined as $\mathcal{R} = 100|N_i - N_o|/N_o$, where N_i is the number of species in mechanism i, and N_o is the number of species in the starting mechanism. $\mathcal{R}$ is largely proportional to the CSP error-tolerance value. Generally, the results in the figure indicate a power-law scaling of error with $\mathcal{R}$ for small $\mathcal{R}$, with some species exhibiting 1st-order scaling, while others exhibit nearly 3rd-order scaling. Clearly, different species exhibit different sensitivity to mechanism simplification. Moreover, the results indicate non-monotonic error behavior at large $\mathcal{R}$, at least for some species, e.g. CH_2 $CH_2(S)$, HCO, H_2O_2, and OH. This behavior is of interest, and is the subject of ongoing work. Finally, note that errors in some species are observed to grow to significant levels for high degrees of reduction. It is, of course, not surprising to find that some species which are not part of the initial kernel set may exhibit such large errors. These errors can be reduced by including the desired species in the initial kernel species set.

We note that the relationship between the specified CSP error tolerance and the observed *a posteriori* error in the flame solution is of general interest. The CSP error tolerance is relevant to the dynamical behavior of a particular mechanism, and as such is not a strict error bound on the flame solution. On the other hand, as the results suggest, the two are clearly correlated, even for non-kernel species. Clearly, however, the form of this correlation is not in a manner that is universal for all species. More work is needed to evaluate the various facets of this relationship for a particular starting mechanism.

4. Conclusions

We have discussed aspects of the application of high-order block-structured AMR in reacting flow computations. We examined the use of explicit stabilized time integration of coupled diffusive-convective terms based on a modified RKC construction. We presented results exhibiting the

dependence of the number of RKC stages on the grid Peclet number, and illustrated fourth-order coupled AMR reaction-convection-diffusion comnputations using this construction in 2D. We also cited the first release of our high-order AMR library, which establishes a key element of our dissemination effort for this work. Focusing on premixed methane-air flame computations, we also studied *a posteriori* errors in flame species using a range of simplified chemical mechanisms developed based on computational singular perturbation (CSP) theory. Results indicate definite correlation of flame-solution accuracy with the CSP importance index error threshold, for a wide range of species in the flame. Work is in progress to further explore these results under broader ranges of mixture conditions.

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