Efficient numerical methods for the simulation of flow reactors

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ABSTRACT

We describe recent developments in the design of numerical methods for the simulation of heat transport and chemical processes in flow reactors. For viscous flow in the low-Mach-number regime, we use a stable finite element method for discretizing the compressible Navier-Stokes equations which is oriented by the incompressible limit case. The flow and chemical equations are solved as a fully coupled system by an adaptive defect correction method. This approach is illustrated by several examples of flow reactors including a case with steep temperature gradients. The efficiency and predictive capability of our codes are verified by comparison with traditional ad-hoc methods and experimental data.

Keywords: flow reactors, finite elements, low-Mach number flow, hp-method

1 INTRODUCTION

1.1 The Mathematical model

The reactive flows considered in this paper are described by the compressible Navier-Stokes equations in the "low-Mach-number" approximation. Here, the pressure p is split into a thermodynamic part P_{th}, which is constant in space, and a hydrodynamic part p_{hyd}, which alone is used in the equation of state. We denote by $D_t := \partial_t + v \cdot \nabla$ the material derivative. The whole system of conservation equations written in primitive variables (p, v, T) takes the following form:

- **mass:**

$$T^{-1} D_t T - \nabla \cdot v = P_{th}^{-1} \partial_t P_{\rm th},$$

- **momentum:**

$$\rho D_t v - \nabla \cdot (\mu\sigma) + \nabla p_{hyd} = \rho f_e,$$

- **energy:**

$$\rho c_p D_t T - \nabla \cdot (\lambda \nabla T) - \partial_t p_{hyd} - \mu\sigma : \nabla v = \partial_t P_{th},$$

- **species:**

$$\rho D_t w_i + \nabla \cdot j_i = f_i(T, w), \quad i = 1, \cdots, n_s,$$

where $\sigma := (\nabla v + \nabla v^T) - \frac{2}{3}(\nabla \cdot v)\mathbf{1}$ is the *shear-stress tensor*, c_p the *heat capacity*, λ the *heat conductivity*, f_e *volume force* and μ the *dynamic shear viscosity*. For the chemical reactions, we consider the formulation in mass fraction w_i of the n_s chemical species. For the diffusion fluxes j_i, we only consider the mass diffusion described by Fick's law. Diffusion due to pressure gradient and thermo-diffusion (Soret-effect) are neglected (see [8] for a motivation of this simplification). The source terms are denoted by $f_i(T, w)$. The equation of state is written in the form

$$\rho = P_{th}/RT, \tag{1}$$

The time derivative of P_{th} is obtained by first averaging the continuity equation in space and then substituting $D_t T$ by using (1), whereas the heating due to p_{hyd} and $\mu\sigma : \nabla v$ is neglected. This leads to a linear scalar ODE of the form:

$$\partial_t P_{th} = F(v, T)\, P_{th}, \tag{2}$$

where $P_{th}(0) = P_0$ is a given initial value (see [7] for more details). Our numerical approach is based on a variational formulation of the set of conservation equations. It is obtained by multiplying the equations by appropriate test functions $\{\chi, \psi, \pi\} =: \phi$ and integrating over the domain Ω. In the diffusion terms integration by parts is used. Neumann-type boundary conditions are implicitly represented by the variational formulation, while Dirichlet boundary conditions have to be explicitly imposed on the solution. Then the variational problem then reads in short: Find $u(t) := \{T(t), v(t), p_{hyd}(t)\} \in V + u_b$, such that $u(0) = u_0$ and

$$(Q\partial_t u, \phi) + a(u; \phi) = F(\phi) \quad \forall \phi \in V\,. \tag{3}$$

Here, Q is a suitable coefficient matrix multiplying the time derivatives, $a(\cdot;\cdot)$ is the semi-linear form corresponding to the variational formulation of the stationary terms of the conservation equations and V is an adequate function space. The right-hand side $F(\cdot)$ contains the *slave variable* P_{th} given by the relation (2), while ρ is determined through the modified gas law (1). The term u_b represents prescribed boundary data.

1.2 Finite Element Discretization

Our Navier-Stokes solver uses a fully implicit approach for solving the "low-Mach-number" approximation of the compressible Navier-Stokes system (3) (for a detailed description we refer to [2], [3]).

The Galerkin finite element method is defined on quadrilateral/hexahedral meshes $\mathcal{T}_h = \{K\}$ covering the domain $\bar{\Omega}$. The trial and test spaces $V_h \subset V$ consist of continuous, piecewise polynomial vector functions (so-called Q_p-elements) for all unknowns, i.e., $p|_K \in Q_s$, $v|_K \in Q_r$, and $T|_K \in Q_r$,. Here, Q_k is the space of (isoparametric) tensor-product polynomials of degree k. The corresponding discrete trial and test spaces are denoted by $V_h \subset V$. In order to facilitate local mesh refinement and coarsening, we allow the cells in the refinement zone to have nodes which lie on faces of neighboring cells (Fig. 1). The degrees of freedom corresponding to such "hanging nodes" are eliminated from the system by interpolation enforcing global conformity (i.e., continuity across interelement boundaries) for the finite element functions.

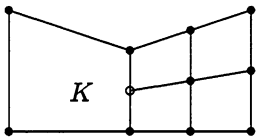


Figure 1: Mesh patch with a "hanging node"

We note that by choosing the trial functions for the pressure of (sufficiently) lower degree than of those for the velocity the form $a(\cdot;\cdot)$ is stable on the discrete spaces V_h (uniformly in h), i.e., it satisfies the uniform "Babuska-Brezzi inf-sup-stability" condition. This particularly guarantees a stable approximation of the pressure. For the elements $Q2/Q1$ and more generally Q_p/Q_{p-2}, for $p > 2$, this condition is fulfilled. In the case of equal-order trial functions for v and p, e.g., the popular Q_1/Q_1-ansatz, the scheme requires "pressure stabilization". In addition, the dominant convection is stabilized by the usual SUPG approach "streamline diffusion" (see e.g. [6] for more details).

2 SOLUTION APPROACH

The solution process consists of several nested loops. The outermost loop is an implicit time iteration for nonstationary reactive flows. The nonlinear system arising on each steps is solved by means of a quasi-Newton iteration. Each Newton step requires the solution of a linear system. The Jacobian is explicitly derived from the finite element discretization and has a sparse structure. We consider the preconditioned GMRES (Generalized Minimal Residual) method which applies to nonsymmetric and indefinite matrices in order to solve this linear system. The preconditioner is based on multigrid techniques. This is a key feature of the overall solution process since for the purely diffusive case (e.g. Laplace equation) its CPU costs grow only linearly with the number of unknowns in contrast classical purely algebraic approaches. The main idea of the multigrid method is based on the ability to derive relatively cheap methods so called *smoothers* that can damp high frequency modes of the error on a given grid. Considering the hierarchy of all available grids allow to damp all frequencies of the error. In our approach, we consider blocking techniques for the smoothers based on the resolution of a local problem. The overall solution process is then as follow:

- Implicit time stepping.
- Nonlinear steps.
  - Quasi-Newton (line search Strategy).
- Linear solver.
  - Iterative solution by GMRES.
  - Preconditioning by multigrid methods.
  - Smoothers based on blocking techniques.

In computing really nonstationary flows the full coupling may be lifted by "operator splitting".

3 APPLICATIONS

3.1 Preamble

The goal of this section is to validate the previously described solution approaches considering applications with increasing difficulties. We emphasize the efficiency and predictive capabilities of the resulting codes by comparison with traditional methods as well as experimental data. We concentrate on cases which require a fully implicit solution process due to stiff coupling of unknowns. The considered applications as well as their intrinsic difficulties are depicted in the following:

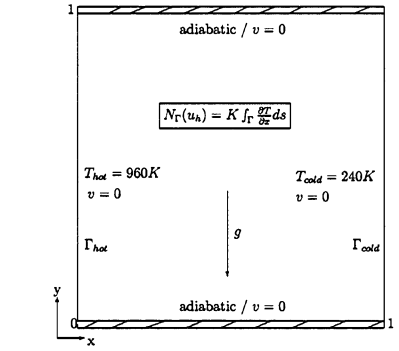
- **Heat driven cavity**: The difficulty of this problem is the large temperature gradient which leads to a highly nonlinear behavior including sharp bound ary layers. This problem has been proposed as benchmark in order to validate and compare the performances of different codes. This problem does not involve chemical reactions and allows therefore to enlight the capabilities of the flow components in the low-Mach-number regime of the code.
- **Chemical reaction in a mixing unit (test case)**: The goal of this academic problem is to compare the efficiency of a general purpose ad-hoc simulation software with our codes for a simple configuration of reactive flow. Besides the large differences in computation time, it is important to notice the scaling of the CPU time with the number of unknowns.

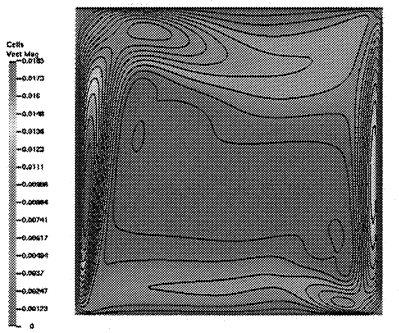
- **CARS experiment**: The CARS (Coherent Antistokes Raman Spectroscopy) experiment involves high efficiency standards for the simulation in order to make parameter identification feasible in a reasonable time. We illustrate the capabilities of the *hp-finite element method* in that context.

If not explicitly mentioned, the presented examples are computed with the C++ FEM code HiFlow (see [5]).

3.2 Heat driven cavity

The discretization described above has first been tested-for various model problems of interior flows including a new heat-transfer benchmark "temperature-driven cavity" for Ra = 10^6 comprising large temperature gradients (Fig. 2).





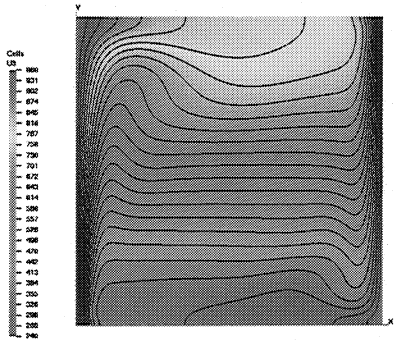


Figure 2: Configuration of the "temperature-driven cavity" flow benchmark (top), velocity norm (middle) and temperature isolines (bottom).

Table 1 shows some representative results. It turns out that higher-order finite elements have good potential for accurately computing interior flows even in the presence of strong layers. The solution of the algebraic problems is the bottleneck in using higher-order finite elements within implicit flow solvers. This problem is tackled by using hierarchical multilevel techniques with blockwise smoothers.

FE ansatz	#cells	#dofs	# entries	CPU time
Q2/Q1	16,384	214,788	12,069,136	2,434 s
Q3/Q1	4,096	115,972	9,558,544	973 s
Q4/Q2	1,024	54,148	6,587,904	602 s
Q5/Q3	256	22,084	3,707,536	633 s

Table 1: Accuracy of higher-order finite elements for solving the "temperature-driven cavity" problem with Ra = 10^6 and Sutherland's law for the viscosity (error $\sim 1\,\%$, reference Nusselt number $N_\Gamma = 8.6866$).

3.3 Chemical reaction in a mixing unit

3.3.1 Academic test case

The flow domain is a rectangular channel of length $L = 2cm$ and width $l = 0.5mm$. At the inlet the parabolic velocity profile (with $1m/s$) and a constant temperature $T_{in} = 273.15K$ are prescribed. The side walls are assumed to be adiabatic. The chemical reaction contains 6 species with 2 exothermal reactions at strongly differing time scales. The goal is to compute the maximal temperature in the resulting stationary flow. This very simple test problem leads to very different CPU requirements by the codes considered. We particularly compared the commercial general purpose code CFX-4 against the academic research code GASCOIGNE (see [1]) developed at the university of Heidelberg. The computations have been done on a HP-9000 with 240 MHz and a SUN UltraSparc 333 MHz, respectively.

# unknowns	CFX-4	Gascoigne	T_{max}
2000	30 min.		1019.93
2500		2 min.	1022.17
5000	240 min.		1019.91
9500		6 min.	1019.29
37000		14 min	1018.41

Table 2: Comparison of the CPU-time (in sec.) needed for the test case 3.3.1

3.3.2 CARS (Coherent Antistokes Raman Spectroscopy) experiment

The goal of this experiment is the measurement of elementary relaxation processes and vibrational energy transfer in collisions of vibrationally excited hydrogen and deuterium molecules. Spatially resolved axial and

radial concentration profiles of both species are obtained at room temperature using *coherent anti-stokes Raman spectroscopy (CARS)* in flow reactor (see Figure 3). The wall deactivation probability and the thermal rate constants for vibrational energy transfer of the reactions

$$\begin{aligned} H_2^{(\nu=1)} &\rightarrow H_2^{(\nu=0)}, \\ H_2^{(\nu=1)} + D_2^{(\nu=0)} &\rightarrow H_2^{(\nu=0)} + D_2^{(\nu=1)}, \end{aligned}$$

respectively, are derived from the detailed numerical simulation through an optimization procedure (see [4] for a more detailed description). In the following, we concentrate on a single step of this optimization scheme which corresponds to the simulation of the overall reactive flow for given rate constants. The considered reaction mechanism includes 32 reactions and involves 9 species.

A comparison is made on computations on tensor product meshes which are globally refined with varying polynomial degrees *(p-method)* and on locally refined grids with varying polynomial degrees *(hp-method)* (see Table 3). The criteria for the local refinement of the grid are based on techniques relying on the a posteriori error estimation corresponding to a sensitivity analysis of the problem with regard to a given functional (see [2]). In our case this "error-control" functional is naturally chosen as the mean value of the error along the cross section of the measurement line. The resulting grid is shown in Figure 4. The computations described in Table 3 have been done on a PC (Pentium III, 500 MHz).

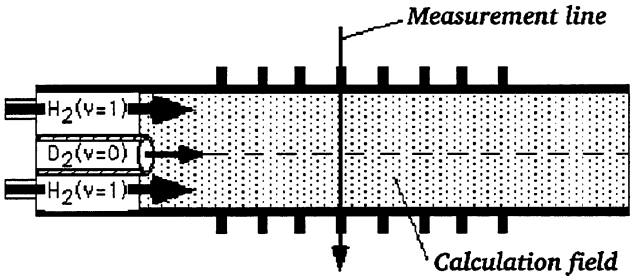


Figure 3: Configuration of the flow reactor

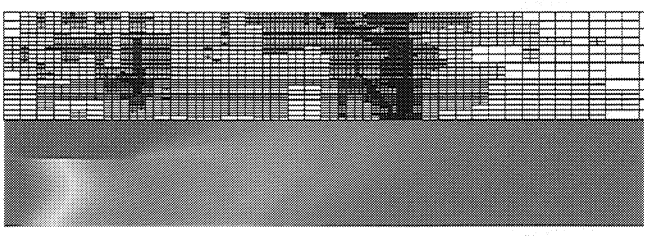


Figure 4: Mass fraction of $D_2^{(\nu=1)}$ in the flow reactor on an adaptively refined mesh.

Refinement	FEM ansatz	#dofs	CPU time
global	Q2/Q1	380,432	160 min.
global	Q3/Q1	148,403	
global	Q4/Q2	102,506	
hp	hp_{Q2-Q8}	43,404	48 min.

Table 3: Needed number of degrees of freedom in order to reach an error smaller than 10% compared to experimental data with regard to the concentration of $D_2^{(\nu=1)}$ on the measurement line.

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