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A NEW, RELIABLE FAMILY OF SPLIT-OPERATOR METHODS FOR COMPUTING REACTING FLOWS

J. B. GREENBERG*

Department of Aeronautical Engineering, Technion-Israel Institute of Technology, Haifa, Israel

SUMMARY

A new, reliable family of split-operator methods is presented for the computation of reacting flows. The methods may be inconsistent or consistent depending upon their applicability. Analyses are carried out for flow combusting according to general multicomponent, multireaction linear or non-linear chemical kinetic models, and conditions for guaranteed stability/convergence are established. A special chemical splitting parameter can also be embedded in the methods enabling *accelerated* convergence of an iterative finite-difference solution. Computed results highlight the capabilities and properties of the methods.

KEY WORDS Finite Differences Split Operators Reacting Flow.

1. INTRODUCTION

The problem of the stiffness of the equations governing reactive systems, whether static or dynamic, has been known since the first computations of laminar flames were performed some thirty years ago.[1] The wide spectrum of reactive collision times, that enters the chemical production terms via the kinetic rate constants, produces a set of, say, n eigenvalues whose orders of magnitude usually obey the following inequality:

$$\min_{1\leq i\leq n} \left|\operatorname{Re}(\lambda_i)\right| \Big/ \max_{1\leq i\leq n} \left|\operatorname{Re}(\lambda_i)\right| \ll 1 \tag{1}$$

Unfortunately, it is generally the eigenvalue possessing the *smallest* real part that sets the bound for the *maximum* time step for numerical integration, even when those terms associated with this eigenvalue are of little physical importance. For *static* reacting systems (or one-dimensional dynamic problems) the governing ordinary differential equations can be reliably solved using the, now standard, algorithm of Hindmarsh[2] based on Gear's method.[3] Alternative o.d.e. solvers are discussed elsewhere.[4–6] For reacting *flow systems* the complex way in which the conservation equations for momentum, energy, species and mass are linked only serves to accentuate the stiffness difficulty. A variety of such problems have been tackled using inconsistent operator-splitting techniques[7–13] in which the more slowly varying portions of the equations are treated using a large time (or space) step, whereas the stiffness-causing chemical kinetics contributions are tackled *separately* using an appropriate smaller integration step. (Some theoretical justification for this kind of approach is outlined by Yanenko[14]) The term 'inconsistent' is applied, since not all the terms of the original

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* Currently on sabbatical leave at Division of Applied Sciences, Pierce Hall, Harvard University, Cambridge, Massachusetts 02138, U.S.A.

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differential equations appear at each stage of the splitting. Under circumstances in which there is *not* strong coupling between the various participating physico-chemical processes their separate numerical treatment is valid and, indeed, efficient. However, as has been pointed out,[15] there *do* exist certain problems in which the coupling *is* strong, so that inconsistent splitting becomes invalid. In such cases *consistent* splitting has been performed, with the difficult chemical source terms being linearized about the species concentrations of the previous time or iterative step (see Reference[15] for relevant references). Linearization of one form or another seems to work (sometimes with the aid of some manipulation based on physical intuition[16]) but does not appear to be foolproof in terms of stability/convergence.

In the current paper a new generalized family of operator-splitting methods is presented for reacting flows. Members of the family are of either the inconsistent or consistent types. For inconsistent members a convergence/stability analysis is outlined for the case of flow in which the combustion is modelled by a general multicomponent (quasi-) unimolecular set of chemical reactions. The analysis leads to conditions for convergence/stability, and also indicates how accelerated iterative solutions may be obtained by suitable regulation of a special chemical splitting parameter. In addition, for non-linear kinetics the 'correct' form of quasi-linearization of the source terms is deduced so that convergence/stability is *completely ensured* when using consistent splitting. Finally, some computed examples highlight the properties of the family of methods.

2. GOVERNING EQUATIONS AND CHEMICAL KINETICS

The governing species conservation equations for a two-dimensional, possibly turbulent, reacting flow are of the form

$$\frac{\partial m_i}{\partial t} = Cm_i + Dm_i + S_i, \qquad i = 1, 2, \ldots \mathrm{NS} \tag{2}$$

where m_i is the mass fraction of species i, C and D are convection and diffusion operators, respectively, and S_i is the chemical source term for species i. In general, there will be coupling between the NS equations (2), the conservation equations for mass, momentum and energy and any equations that comprise a turbulence model. It is presumed that appropriate boundary and initial conditions are specified on the boundary δB of the region B within which the governing equations are valid. Let there be NR reversible chemical reactions occurring between the NS species of the system:

$$\sum_{j=1}^{\mathrm{NS}} \nu_{ij}\Lambda_j \rightleftarrows \sum_{j=1}^{\mathrm{NS}} \nu^1_{ij}\Lambda_j, \qquad i = 1, 2 \ldots \mathrm{NR} \tag{3}$$

where ν_{ij}, ν^1_{ij} are the stoichiometric coefficients of Λ_j occurring as a reactant or product, respectively, in reaction i. The chemical source terms S_i then assume the form

$$S_i = M_i \sum_{q=1}^{\mathrm{NR}} (\nu^1_{qi} - \nu_{qi}) \left[k_{\mathrm{F}q} \prod_{S=1}^{\mathrm{NS}} \left[\frac{\rho m_S}{M_S}\right]^{\nu_{qS}} - k_{\mathrm{R}q} \prod_{S=1}^{\mathrm{NS}} \left[\frac{\rho m_S}{M_S}\right]^{\nu^1_{qS}} \right] \tag{4}$$

where ρ is the mixture density, M_i is the molecular weight of species i, and $k_{\mathrm{F}q}$ and $k_{\mathrm{R}q}$ are the forward and reverse rate constants of reaction q. The rate constants are generally expressed in Arrhenius form:

$$k_q = A_q T^{\gamma_q} \mathrm{e}^{-(E_q/RT)} \tag{5}$$

where γ_q, A_q are constants, T is the temperature, E_q is the activation energy for the reaction and R is the universal gas constant. If the combustion of the reacting flow system is modelled by a set of quasi-unimolecular reactions occurring between the NS species, then the expressions (3) and (4) simplify to

$$\Lambda_i \rightleftharpoons \Lambda_j, \qquad (i \neq j) \text{ for all } i, j \tag{3a}$$

$$S_i/M_i = -\sum_{j=1}^{\mathrm{NS}} k_{ij} m_i + \sum_{j=1}^{\mathrm{NS}} k_{ji} m_j, \qquad i = 1, 2 \ldots \mathrm{NS} \tag{4a}$$

where k_{ij} is the rate constant associated with the production of Λ_j from Λ_i. Henceforth, a distinction will be made between the non-linear kinetics ((3) and (4)) and the linear kinetics ((3a) and (4a)), which will be referred to as NLK and LK, respectively. If τ_{C} and τ_{D} are characteristic times for the physical processes of convection and diffusion, and τ_q is the reactive collsion time for reaction q, then a typical ordering of times will be

$$\tau_1 < \tau_2 < \tau_3 \ldots \tau_l \ll \tau_{l+1} < \tau_{l+2} < \ldots < \tau_{2\mathrm{NR}} < (\tau_{\mathrm{C}} \text{ or } \tau_{\mathrm{D}}) \tag{6}$$

The wide disparity between the various time (or, equivalently, length) scales is responsible for the stiffness of the governing equations (2), which prevents attainment of a numerical solution by standard finite-difference methods.

3. SPLIT-OPERATOR METHODS

(a) *Linear kinetics and inconsistent splitting*

For simplicity the case of LK will be considered first. Equation (2) can then be written in the general form

$$\frac{\partial \mathbf{m}}{\partial t} = (\mathbf{C} + \mathbf{D} + \mathbf{A})\mathbf{m} \tag{2a}$$

where $\mathbf{m}$ is the mass fraction vector and $\mathbf{C}$, $\mathbf{D}$ and $\mathbf{A}$ are matrices of convection, diffusion and LK, respectively. For clarity the latter matrix's elements are

$$A_{ij} = -\sum_{p=1}^{\mathrm{NS}} k_{ip}\, \delta_{ij} + k_{ji}(1 - \delta_{ij}) \tag{7}$$

with δ_{ij} being the Kronecker delta.

The formal solution of (2a) can be written as

$$\mathbf{m}(t) = \exp\left[t(\mathbf{C} + \mathbf{D} + \mathbf{A})\right]\mathbf{m}(0) \tag{8}$$

whence split-operator approximate methods can be constructed.

The governing principle for constructing *inconsistent split-operator methods* is usually to separate the solution, (8) into several parts, each of which is compatible with its own typical time scale. Here, the splitting will assume a more general appearance so as to encompass the possibility of accelerating convergence to a steady state, as reported recently for the reacting flow context.[17]

Equation (8) is rewritten in the following form:

$$\exp\left[-t(1-\beta)\mathbf{A}\right]\mathbf{m}(t) = \exp\left[t(\mathbf{C} + \mathbf{D} + \beta\mathbf{A})\right]\mathbf{m}(0) \tag{8a}$$

whence the following two-steps are derived:

$$\mathbf{m}^* = \exp\,[t(\mathbf{C}+\mathbf{D}+\beta\mathbf{A})]\mathbf{m}(0) \tag{9a}$$

$$\mathbf{m}(t) = \exp\,[t(1-\beta)\mathbf{A}]\mathbf{m}^* \tag{9b}$$

where β, lying between zero and one, is called the chemical splitting parameter. These two steps provide the basis for the finite-difference formulae by means of which equation (2a) is solved. For example, expanding (9a) and replacing $\mathbf{C}$ and $\mathbf{D}$ by appropriate finite-difference operators, $\bar{\mathbf{C}}$, $\bar{\mathbf{D}}$, leads to the fully explicit scheme for the first step:

$$\mathbf{m}^* = [1+(\bar{\mathbf{C}}+\bar{\mathbf{D}}+\beta\mathbf{A})t]\mathbf{m}(0) \tag{10a}$$

Alternatively, implicit methods can be constructed, one of which has been used in this work:

$$[1-\beta\mathbf{A}t]\mathbf{m}^* = [1+(\bar{\mathbf{C}}+\bar{\mathbf{D}})t]\mathbf{m}(0) \tag{10aa}$$

As for the second stage (9b); it turns out to be convenient to translate it back into differential form:

$$\frac{\mathrm{d}\mathbf{m}}{\mathrm{d}t} = (1-\beta)\mathbf{A}\mathbf{m} \tag{10b}$$

Only stiffness-causing elements are contained in this stage. However, the availability of many stable stiff-o.d.e. solvers renders the stiffness problem unimportant *vis-a-vis* obtaining a numerical solution. Such an o.d.e. solver will take many small time steps to complete one sweep of stage (10b). This can be viewed as resplitting (9b) into many components:

$$\mathbf{m}(t) = \left[\prod_{i=1}^{N} \exp\,[t_i(1-\beta)\mathbf{A}\right]\mathbf{m}^* \tag{9c}$$

where

$$\sum_{i=1}^{N} t_i = t \tag{11}$$

Several points should be noted for the split-operator method consisting of (10aa) and (10b). Unlike the previous chemical splitting scheme suggested by the author[17] the current one is *chemically consistent* at each stage, and, therefore, more accurate. Each value of β defines a member of this new family of methods. The method of Greenberg and Presser[11] is recovered by setting $\beta = 0$. For elliptic governing equations (i.e. when the steady state is presumed from the outset rather than approached asymptotically via the limit $\partial/\partial t \rightarrow 0$) an appropriate choice of t reproduces identically the iterative method adopted in Reference 13, where Hindmarsh's algorithm was used for equation (10b). This simple manner of splitting indicates how complex kinetics can be incorporated with relatively little effort within the framework of *existing* elliptic codes, if required.[11,18] It should be pointed out that, whereas for the parabolic equations the intermediate $\mathbf{m}^*$ can be taken as $\mathbf{m}(t/2)$ so that integration of (10b) proceeds from $t/2$ to t, for the elliptic case the limits of integration are taken as dependent on local characteristic convection and diffusion times. Specifically, it has been previously shown to be correct to integrate (10b) from $t = 0$ to τ where τ is given by[17]

$$\tau = \min\left\{\left|\frac{\Delta x}{u}\right|, \left|\frac{\Delta y}{v}\right|, \left|\frac{\Delta x^2}{D}\right|, \left|\frac{\Delta y^2}{D}\right|\right\} \tag{12}$$

where u, v are velocity components and D is the diffusion coefficient.

Finally, it is pointed out that a similar alternative consistent approach to treating stiffness has been reported[7] using the method of lines. Only the RHS of equation (2a) is discretized leading to a large coupled set of o.d.e.s that can be solved using a stiff-o.d.e. solver. All points in the field are simultaneously coupled but, consequently, computational overheads and time are large.[7] On the other hand, the aforementioned splitting approach (when applicable) has the advantage of necessitating the solution of smaller sets of o.d.e.s (NS equations) *separately* at each point in the field, the coupling to neighbouring points resulting from the first, more rapidly executed, stage of the splitting. For multidimensional problems splitting is computationally preferable.

(b) *Analysis and properties*

Having described the construction and application of the inconsistent splitting methods their special properties will be investigated. In order to gain some insight into conditions for convergence/stability, and to clarify the role of the chemically splitting parameter, β, an analysis of the linear case will be performed.

The general finite-difference formulation of Chien[19] is adopted for the convection and diffusion operators. It is noted that as equations (10aa) and (10b) stand there is coupling between the elements of $\mathbf{m}$ because the matrix $\mathbf{A}$ is fully populated. However, by using a matrix similarity transformation

$$\mathbf{m} = \mathbf{M}\boldsymbol{\eta} \tag{13}$$

the following decoupled finite-difference equation results for η at each point (j, k) in the field:

$$\begin{aligned}[\rho_{jk}\mathbf{I} - \rho^1_{jk}\beta\mathbf{Q}]\boldsymbol{\eta}^*_{j\,k} = \Bigg[& G_k(\delta y)^2\left\{\left(1-\frac{G_jRe_j}{2}\right)\boldsymbol{\eta}_{j+1,k} + \left(1+\frac{G_jRe_j}{2}\right)\boldsymbol{\eta}_{j-1,k}\right\} \\ & + G_j(\delta x)^2\left\{\left(1-\frac{G_kRe_k}{2}\right)\boldsymbol{\eta}_{j,k+1} + \left(1+\frac{G_kRe_k}{2}\right)\boldsymbol{\eta}_{j,k-1}\right\}\Bigg]\end{aligned} \tag{14}$$

where

$$\mathbf{Q} = \mathbf{M}^{-1}\mathbf{AM} \tag{15}$$

is a diagonal matrix with elements q_s that are the eigenvalues of $\mathbf{A}$, G_j, G_k are Chien's decay functions, Re_j and Re_k are cell Reynold's numbers based on local velocity components and

$$[\rho_{jk}, \rho^1_{jk}] = [2[(\delta x)^2G_j + (\delta y)^2G_k], G_jG_k(\delta x\,\delta y)^2Re] \tag{16}$$

In the above the time-step, t, has been chosen so as to enable the elliptic and parabolic cases to be treated together. This is not essential to the analysis. Since ρ_{jk} and ρ^1_{jk} are always positive it is evident that the signs of the eigenvalues of $\mathbf{A}$ are of utmost importance for stability purposes. It can be proved that the *structure of* $\mathbf{A}$ *dictates only non-positive* eigenvalues (see reference 20 for proof). Now, the solution of equation (10b) (in terms of the similarity variable) is

$$\boldsymbol{\eta} = \exp[(1-\beta)\tau\mathbf{Q}]\boldsymbol{\eta}^* \tag{17}$$

Combining (14) and (17) for a complete time step or a single iterative sweep leads to

$$\boldsymbol{\eta}^{n+1} = \exp[(1-\beta)\tau\mathbf{Q}]\mathbf{Y}\boldsymbol{\eta}^n \tag{18}$$

where the elements of $\mathbf{Y}$ are derived from equation (14).

Thus, if the fluid field is given and fixed it can be shown that a sufficient condition for convergence of the split scheme is given by

$$\max_{j,k,s}\left|\exp\left[-(1-\beta)\,|q_s|\,\tau_{jk}\right]\cdot\left\{(\delta x)^2 G_j\left(\left|1+\frac{G_k Re_k}{2}\right|+\left|1-\frac{G_k Re_k}{2}\right|\right)\right.\right.$$
$$\left.\left.+(\delta y)^2 G_k\left(\left|1+\frac{G_j Re_j}{2}\right|+\left|1-\frac{G_j Re_j}{2}\right|\right)\right\}\Big/\Delta_{jks}\right|<1 \qquad (19)$$

where

$$\Delta_{jks}=2\left|(\delta x)^2 G_j+(\delta y)^2 G_k+\frac{G_j G_k}{2}(\delta x)^2(\delta y)^2\beta\,|q_s|\,Re\right|>0 \qquad (20)$$

whence it is deduced that for stability/convergence it is sufficient that

$$\left|\frac{G_j Re_j}{2}\right|<1 \quad \text{and} \quad \left|\frac{G_k Re_k}{2}\right|<1 \qquad (21)$$

which collapses to the well-known cell Reynolds' number limitation if the decay functions equal unity. For upwind differencing the conditions are once again satisfied. Note that if no splitting is made ($\beta = 1$) conditions (21) still apply with the occurrence of enhanced diagonal dominance stemming from the contribution of the chemistry. For split schemes the occurrence of the exponential term from the chemistry, as well as the enhanced diagonal dominance only serves to relax conditions (21) under certain circumstances. Furthermore, it should be noted that it is possible to attain accelerated convergence with the help of the chemical splitting parameter. The functional dependence of the LHS of equation (19) on β is

$$f(\beta)=a\mathrm{e}^{\beta}/(b+c\beta) \qquad (22)$$

A minimum value of f exists in the range $0 \leq \beta \leq 1$ so that appropriate regulation of β can be expected to accelerate convergence of an iterative procedure. Unfortunately, *a priori* estimation of the optimum value of β is time-consuming and difficult, particularly if the NLK case is under consideration. This situation is somewhat analogous to the trial and error determination of an optimal under-relaxation parameter for iterative solution of non-linear systems of equations.

(c) *Nonlinear kinetics and consistent splitting*

The shortcomings of inconsistent splitting methods have been alluded to in the Introduction. For strong coupling between the physico-chemical processes a *consistent splitting scheme* is preferable, in which all processes contribute explicitly to *each* stage of the splitting. For illustrative purposes let $\mathbf{C} \equiv \mathbf{0}$ in equation (8) and suppose D is the Laplacian:

$$D=\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2} \qquad (23)$$

Furthermore, assume NLK so that:

$$\mathbf{A}=\mathbf{A}(\mathbf{m}) \qquad (24)$$

implying that the formal solution, equation (8), is now approximate:

$$\mathbf{m}(t) \simeq \exp\left[t(\mathbf{C}+\mathbf{D}+\mathbf{A}(\mathbf{m}(0)))\right]\mathbf{m}(0) \qquad (8b)$$

A *consistent splitting scheme* will be

$$\exp\left[-\frac{t}{2}\left(\frac{\partial^2}{\partial x^2}+\mathbf{A}\right)\right]\mathbf{m}^* = \exp\left[\frac{t}{2}\left(\frac{\partial^2}{\partial y^2}+\mathbf{A}\right)\right]\mathbf{m}(0)$$

$$\exp\left[-\frac{t}{2}\left(\frac{\partial^2}{\partial y^2}+\mathbf{A}\right)\right]\mathbf{m}(t) = \exp\left[\frac{t}{2}\left(\frac{\partial^2}{\partial x^2}+\mathbf{A}\right)\right]\mathbf{m}^* \tag{25}$$

(In practice, it will be preferable to evaluate $\mathbf{A}$ during the second stage according to $\mathbf{A}(\mathbf{m}^*)$.) It is not difficult to show that if central differences are used for the space differentials the condition for a stable/convergent solution hinges upon the non-positiveness of the eigenvalues of $\mathbf{A}$. Many workers have constructed $\mathbf{A}$ by time linearization of the source terms[7] leading to the Jacobian $\partial S_i/\partial m_j$, whose eigenvalues need not necessarily be non-positive. To *ensure* negative eigenvalues a sort of quasi-linearization will be performed. The argument is based on the LK analysis. As was shown before, the actual structure of (the LK) $\mathbf{A}$ precluded positive eigenvalues. Therefore, for the NLK case $\mathbf{A}$ will be built so as to have the same element-structure as $\mathbf{A}$ of the LK case. For example, consider a single reversible reaction:

$$\Lambda_i + \Lambda_j \underset{k_R}{\overset{k_F}{\rightleftarrows}} \Lambda_p + \Lambda_l \tag{26}$$

The source terms for the four species will be written as

$$\mathbf{Am} = \begin{bmatrix} -k_F m_j & 0 & 0 & k_R m_p \\ 0 & -k_F m_i & k_R m_l & 0 \\ 0 & k_F m_i & -k_R m_l & 0 \\ k_F m_j & 0 & 0 & -k_R m_p \end{bmatrix} \begin{bmatrix} m_i \\ m_j \\ m_p \\ m_l \end{bmatrix} \tag{27}$$

Now, define the following 'constants':

$$K_{il} = k_F m_j, \qquad K_{li} = k_R m_p, \qquad K_{jp} = k_F m_i, \qquad K_{pj} = k_R m_l \tag{28}$$

Then, $\mathbf{A}$ assumes the form:

$$\mathbf{A} = \begin{bmatrix} -K_{il} & 0 & 0 & K_{li} \\ 0 & -K_{jp} & K_{pj} & 0 \\ 0 & K_{jp} & -K_{pj} & 0 \\ K_{il} & 0 & 0 & -K_{li} \end{bmatrix} \tag{29}$$

which can be thought of as derived from the following pseudo-unimolecular reaction set:

$$\Lambda_i \underset{K_{li}}{\overset{K_{il}}{\rightleftarrows}} \Lambda_l\,; \qquad \Lambda_p \underset{K_{jp}}{\overset{K_{pj}}{\rightleftarrows}} \Lambda_j \tag{30}$$

$\mathbf{A}$ then has non-positive eigenvalues as required. Construction of $\mathbf{A}$ in this fashion is *always possible*, irrespective of the number of participating reactions. Hence, stability/convergence of the consistent splitting scheme derived from (25) can be guaranteed. The chemical splitting parameter can also be inserted into these schemes to accelerate convergence, if required.

(d) *LK and consistent splitting*; *NLK and inconsistent splitting*

The analysis of the previous section simplifies if linear kinetics are used, since $\mathbf{A}$ (which remains constant now) has non-positive eigenvalues. For NLK with an inconsistent splitting scheme such as (10aa)+(10b) the following line of reasoning applies. Suppose all the chemistry is delegated to the second stage ($\beta = 0$) and a stiff-o.d.e. solver is used. It is safe to say that

$$\boldsymbol{\eta} = \mathbf{P}\boldsymbol{\eta}^* \tag{31}$$

describes this stage with

$$\|\mathbf{P}\| < 1 \tag{32}$$

for some norm. Combining this with conditions (21) (that stem from the first stage of the splitting) implies, once again, that stability/convergence can be made inevitable. In fact, this provides a rational explanation for the successful use of all split-operator methods of this nature.[7–13]

(e) *Truncation errors, order of splitting and iterative convergence criteria*

The local order of accuracy of both the inconsistent and consistent splitting schemes is that of the worst time and spatial approximations used. Thus, typically, for forward time and central space differencing the truncation error will be $O(\Delta t, \Delta x^2, \Delta y^2)$. The differences between the various splitting schemes are expressed via the *coefficients* of Δt, Δx^2, Δy^2. If the chemical splitting parameter β is also used the coefficients are similarly affected and become β-dependent. Proof of convergence (in the sense of the behaviour of the difference equations as Δt, Δx, $\Delta y \rightarrow 0$) is straightforward for linear kinetics only. Numerical experiments seem to indicate that taking a finer mesh and/or a smaller time step causes 'convergence' to some limiting solution for non-linear kinetics (see later). The *order* of execution of the splitting can be of great importance. It is *irrelevant* for LK with both consistent and inconsistent splitting, and NLK with consistent splitting. (Parenthetically, it is noted that it is preferable in the latter case to iterate the second stage for each time step; convergence occurs generally within two or three iterations.) However, care must be exercised for NLK with inconsistent splitting, for reversing the order of execution can produce different results. The extent of such a discrepancy depends on how much more dominant the convection–diffusion process is than the chemistry, or vice versa. For large predominance (several orders of magnitude) the discrepancy is generally negligible, since the perturbation caused by the less dominant process is small. When there is stronger coupling between the processes the perturbations are large so that inconsistent splitting may produce the wrong solution. It is recommended to monitor the changes due to the various processes and to decide whether inconsistent or consistent splitting should be applied in accordance with some relative magnitude criterion. This procedure optimizes, in some sense, both execution time and accuracy of the solution. For elliptic problems iterative convergence should be measured (for inconsistent splitting) in terms of both the increments induced due to each split stage *separately* and that resulting from application of both the stages.

4. COMPUTED RESULTS

In order to investigate the properties of the new family of methods some test calculations were performed, with a variety of linear and non-linear kinetic schemes.

(a) *Linear kinetics—reacting Couette flow*

The problem at hand is illustrated in Figure 1. There is injection of the reactants at the

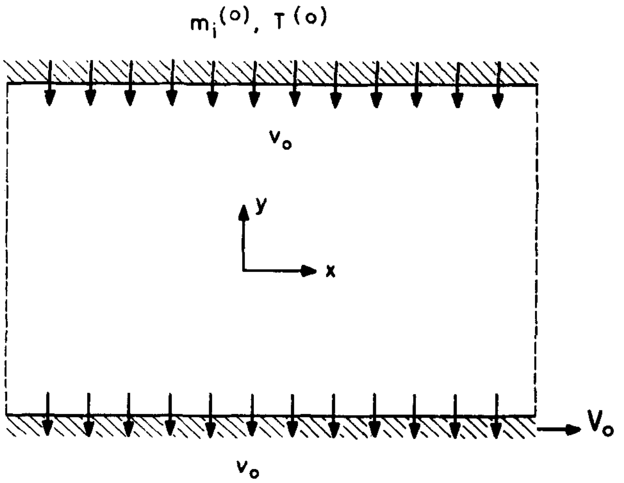

Figure 1. Couette flow with injection and suction

stationary wall and equal suction at the horizontally travelling wall. The pressure gradient is zero. Assuming Prandtl and Schmidt numbers of unity produces a trivial constant enthalpy solution. The non-dimensionalized governing equations were:

$$u\frac{\partial\boldsymbol{\phi}}{\partial x}+v\frac{\partial\boldsymbol{\phi}}{\partial y}=\frac{1}{Re}\left[\frac{\partial^2\boldsymbol{\phi}}{\partial x^2}+\frac{\partial^2\boldsymbol{\phi}}{\partial y^2}\right]+\left(\frac{L_0}{V_0}\right)\mathbf{S} \tag{33}$$

where

$$\boldsymbol{\phi}^{\mathrm{T}}=(u, v, m_1, m_2, \ldots m_{\mathrm{NS}}) \tag{34}$$

and

$$\mathbf{S}=(0, 0, S_1, S_2 \ldots S_{\mathrm{NS}}) \tag{35}$$

Boundary conditions were:

$$\begin{array}{llll} u=0, & v=-v_0, & m_i=m_i^{(0)}, & \text{at} \quad y=h \\ u=V_0, & v=-v_0, & \partial m_i/\partial y=0, & \text{at} \quad y=0 \end{array} \tag{36}$$

Table I. Chemical kinetic models and data for reacting Couette flow

Model	Reactions	Rate constants data ($k=\bar{A}\exp(-E/T)T^{\bar{\beta}}$)		
		$\bar{A}$	E	$\bar{\beta}$
1. Linear (hypothetical) case 1.	$A\rightarrow B$	10^3	0	0
	$B\rightarrow A$	10^{-1}	0	0
	$B\rightarrow C$	10	0	0
	$C\rightarrow B$	10^{-6}	0	0
	$A\rightarrow C$	10^4	0	0
	$C\rightarrow A$	10^{-2}	0	0
2. Non-linear (ozone decomposition)*	$O_3+M\rightarrow O_2+O+M$	$9{\cdot}9\times10^{14}$	+11,350	0
	$O+O_2+M\rightarrow O_3+M$	$7{\cdot}4\times10^{12}$	1050	0
	$O+O_3\rightarrow 2O_2$	$1{\cdot}2\times10^{13}$	−2395	0
	$O+O+M\rightarrow O_2+M$	1·4	−170	−1

* Data taken from Reference 23

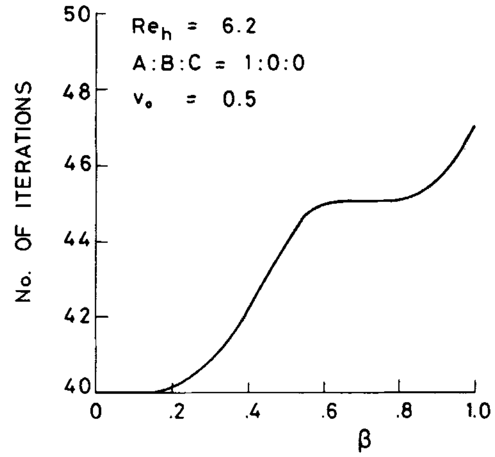

Figure 2. Effect of chemical splitting parameter on convergence rate—linear kinetics

At the vertical boundary conditions of periodicy were imposed. Data relevant to the chemical model are summarized in Table I.

For the inconsistent splitting methods the solution of the second stage was performed using the IMSL adaptation of Hindmarsh's Gear-type algorithm. This method was chosen simply because of its availability. Clearly, *any* stable efficient stiff-o.d.e. solver could have been adopted. The relative error bound supplied for integration was the most liberal one recommended, 10^{-3}. A sparse uniform mesh 5×11 covered the solution region—mesh adaptation was not examined here. For all cases computed convergence was attained (relative error of 5×10^{-4}; see previous section on convergence testing) *irrespective of the initial guesses* (which had simply to obey mass conservation) and the value of the chemical splitting parameter. In Figure 2 a typical curve showing the dependence of the convergence rate on β is shown. The optimal values of β lie in the range $[0, 0{\cdot}25]$ approximately. The number of iterations has been used as a measure of merit rather than CPU time since integration by Gear's method may be overcautious. (Actually, for the problem at hand, CPU time was nearly a linear function of the number of iterations.) The effect of the splitting

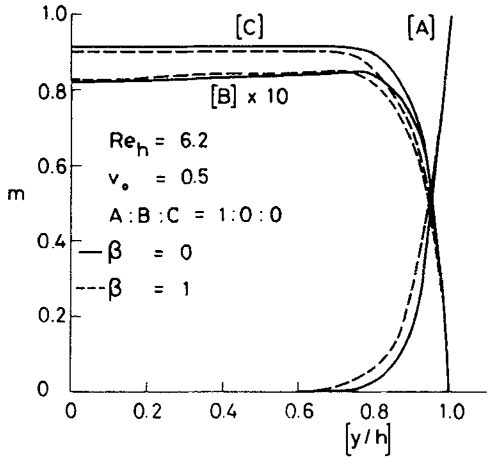

Figure 3. Concentration profiles for reacting Couette flow—linear kinetics

parameter on the concentration profiles is illustrated in Figure 3, where the curves computed using the limiting values of β are shown. The discrepancy stems from the different coefficients in the truncation errors and the sparsity of the mesh. Calculations using more dense meshes of 5×22 and 5×33 virtually annihilated the discrepancy indicating that splitting was valid for the problem at hand. The overall qualitative picture in Figure 3 is consistent with the relative sizes of the rate constants producing and depleting the various species (see Table I).

(b) *Non-linear kinetics—inconsistent splitting*

Two problems involving non-linear kinetics were tackled using inconsistent splitting. The first was Couette flow with ozone decomposition. Relevant data are shown in Table I. The second involved combustion of N_2–O_2 in a 'hypersonic' boundary layer. The outer edge of the layer lies in the region in the front of the shock wave produced, for example, when a blunt-nosed space vehicle re-enters the Earth's atmosphere. The high temperatures experienced lead to the dissociation of N_2 (and O_2). In turn, the heat of reactions in the boundary layer adhering to the nose of the vehicle may produce large temperature gradients and, hence, additional heat flux to the vehicle thus threatening its structural integrity. The governing equations and a thorough study of the effects of the finite chemistry will be given elsewhere.[21] Here it suffices to say that a similarity transformation is applied rendering the governing parabolic equations elliptic. The chemistry is described using the data shown in Table II. A five species, twelve step kinetic model is applied, and it is presumed that equal concentrations of N_2 and O_2 (0·75 and 0·25, respectively) enter the boundary layer from its outer edge and (by injection) from the nose of the vehicle. The fixed ratio between the temperature at the wall and that far from the wall is 0·25. For the computations the matrix **A** was held constant for the first stage of splitting but allowed to vary during the second stage of o.d.e. integration. The temperature was held constant throughout each complete iteration. For the problems examined this did not significantly alter the convergence characteristics. If **A** was constructed 'correctly' (as in Section 3(c)) liberal initial guesses could be supplied. Taking **A** as the Jacobian necessitated more 'correct' initial guesses, similar to the procedure

Table II. Chemical kinetic model and data for reacting hypersonic boundary layer

		Rate constants data ($k=\bar{A}\exp(-E/T)T^{\bar{\beta}}$)		
	*Reactions**	$\bar{A}$	E	$\bar{\beta}$
1.	$O_2+M\rightarrow 2O+M$	$9{\cdot}8\times10^{24}$	−59, 380	−2·5
2.	$2O+M\rightarrow O_2+M$	$4{\cdot}7\times10^{23}$	0	−2·5
3.	$N_2+M\rightarrow 2N+M$	$3{\cdot}7\times10^{21}$	−113, 200	−1·6
4.	$2N+M\rightarrow N_2+M$	3×10^{14}	−500	0
5.	$NO+M\rightarrow N+O+M$	$2{\cdot}3\times10^{17}$	−74, 900	−0·5
6.	$N+O+M\rightarrow NO+M$	$6{\cdot}4\times10^{16}$	0	−0·5
7.	$O_2+N\rightarrow NO+O$	$6{\cdot}4\times10^{9}$	−3150	1
8.	$NO+O\rightarrow O_2+N$	$1{\cdot}5\times10^{9}$	−19, 500	1
9.	$N_2+O\rightarrow NO+N$	$7{\cdot}6\times10^{13}$	−38, 000	0
10.	$NO+N\rightarrow N_2+O$	$1{\cdot}6\times10^{13}$	0	0
11.	$N_2+O_2\rightarrow NO+NO$	$6{\cdot}3\times10^{13}$	−55, 200	0
12.	$NO+NO\rightarrow N_2+O_2$	$1{\cdot}3\times10^{14}$	−38, 060	0

* Data culled from References 24 and 25

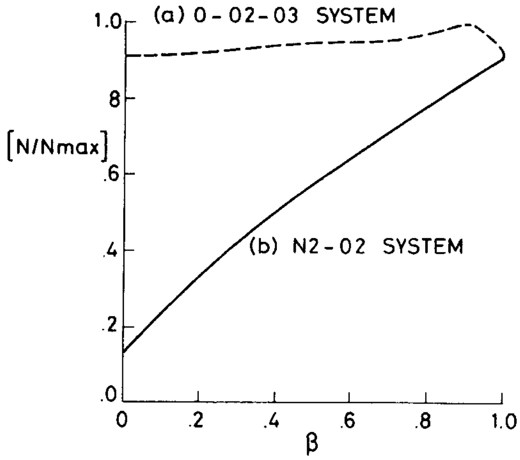

Figure 4. Effect of chemical splitting parameter on convergence—non-linear kinetics: (a) reacting Couette flow, (b) reacting hypersonic boundary layer

described elsewhere.[11] The effect of the splitting parameter on convergence for both non-linear problems is shown in Figure 4. For the ozone-decomposition combustion (unburnt gas ratios $O:O_2:O_3 = 0{\cdot}0:0{\cdot}6:0{\cdot}4$) the complete split scheme was preferred. Interestingly, the worst value of β is 0·9, convergence being 10 per cent slower than the optimum. For the N_2–O_2 system the completely split scheme is again optimal, this time being 770 per cent faster than the regular unsplit scheme! In fact, the dependence of the convergence rate on β appears to be virtually linear for this case.

The mass fraction profiles of the five reacting species are illustrated in Figure 5 and indicate the correct anticipated behaviour. In the thin reaction zone as much as 5 per cent of atomic oxygen is produced and 3 per cent of nitric oxide. The effect of this intense reaction region on the heat flux to the vehicle can be deduced by comparing the temperature gradients obtained with and without chemical reaction ((f) and (g)). The value of $(dT/d\eta)_{\eta=0}$

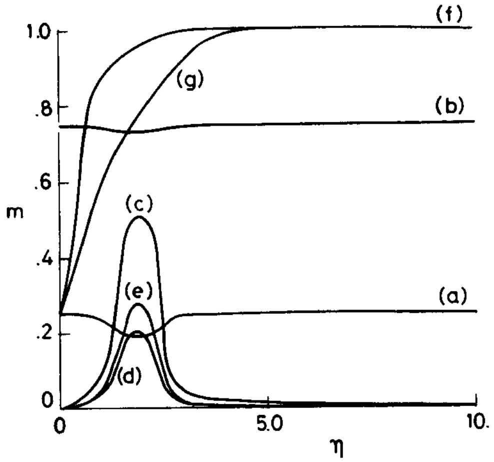

Figure 5. Concentration and normalized temperature profiles in reacting hypersonic boundary layer; $\beta = 0$; (a) O_2, (b) N_2, (c) $O\times10^{+1}$, (d) $N\times10^2$, (e) $NO\times10^2$, (f) T/T_e, (g) T/T_e with no chemical reactions

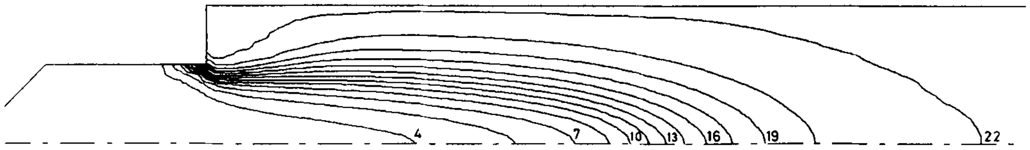

Figure 6. Temperature distribution in axisymmetric combustion chamber (10^2K)

increases twofold due to dissociation of N_2 and emphasizes the importance of accurate modelling of the physics in this context.

(c) *Non-linear kinetics—consistent splitting*

Finally, a more complex problem of reacting flow in a combustion chamber was solved using a consistent splitting scheme. The non-linear chemical kinetics and relevant steady-state equations are reported elsewhere.[22] The problem had been previously solved using a tiring trial-and-error relaxation-type method for the chemical source terms, but involving *no splitting*. The use of the method of Section 3(c) with the 'correct' quasi-linearization of $\mathbf{A}$ enabled the entire solution to be computed without any trial-and-error in about 70 per cent of the CPU time previously required. The solutions were identical to those previously computed. Temperature contours are shown in Figure 6.

5. CONCLUSIONS

A new (chemically consistent) family of split-operator methods has been presented for computing reacting flows. Both inconsistent and consistent splitting is allowed. An analysis has shown conditions for convergence/stability for both linear and non-linear kinetics. In fact, for non-linear kinetics it is possible to construct a quasi-linearized source term matrix with desirable convergence/stability properties. Inclusion of a special chemical splitting parameter can enhance the convergence rate. Computed results indicate the usefulness and reliability of the new methods.

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