# Numerical Study of the Interaction of Fast Chemistry and Diffusion

G. R. Otey*
*Sandia Laboratories, Albuquerque, N. Mex.*
and
H. A. Dwyer†
*University of California, Davis, Calif.*

A model problem that contains the essential features of most combustion processes has been formulated and used to characterize the efficiency of five numerical solution procedures. Chemistry is modeled by both one- and two-step Arrhenius-type reactions, which allows the effect of stiffness on the efficiency of the methods to be assessed parametrically. The numerical schemes evaluated include two methods of lines techniques, two schemes which use pointwise integration of reaction terms in conjunction with different diffusion models, and a locally linearized block tridiagonal, implicit procedure. The latter technique with fourth-order spatial differences has been found to be most efficient over a wide range of reaction rates including cases with considerable stiffness. Combination of this method with adaptive gridding schemes and extension to two-dimensional problems of combustion in a gaseous medium are discussed qualitatively.

## Introduction

**I**N recent years several one-dimensional, time-dependent techniques have been developed for the purpose of analyzing flame propagation.[1-7] In some, a transformation to Lagrangian coordinates is used essentially to eliminate the continuity equation from the solution process, and the momentum equation is eliminated by the assumption of uniform, quasisteady pressure throughout the combustion chamber. While this procedure is convenient in certain one-dimensional cases, the extension to more complex geometries appears difficult. Additionally, it is unsatisfactory for modeling phenomena that take place on acoustic time scales, such as ignition and transition to detonation. The models developed by Boni et al.,[5] Westbrook,[6] and Dwyer and Sanders[7] are somewhat more comprehensive from the standpoint of fluid dynamics in that the equations of continuity, momentum, and energy are solved along with the species equations for either global or two-step chemical kinetics. More recently, two- and three-dimensional models have been presented by Boni et al.,[8] Butler and O'Rourke,[9] and Griffin et al.[10]

For one-dimensional problems, selecting a highly efficient solution technique has not been essential. However, as more complex chemical kinetics and turbulence models are considered, better solution procedures will become more important. Moreover, efficiency in one-dimensional models is important in that the procedure used in many multidimensional schemes (e.g., approximate factorization, alternating-direction implicit methods) is to reduce the problem to a sequence of one-dimensional calculations. More efficient numerical methods will be essential in the development of two- and three-dimensional models. The computation times for the previously mentioned models are moderate only because simple algebraic turbulence models and/or global chemical kinetics are employed.

In addition to the need for developing better numerical methods, a systematic way of assessing their relative efficiency is also needed. Such an assessment should be carried out over a variety of reaction rates, different ratios of reaction zone thickness to chamber dimensions, and with varying degrees of stiffness. Comparisons using two- and three-dimensional test problems would be very expensive. For this reason a one-dimensional model (test) problem has been developed for use in this investigation.

In selecting a model problem it is highly desirable to choose one which is as simple as possible consistent with retaining the essential features affecting computational efficiency. The problem chosen retains the exponential temperature dependence of Arrhenius-type reactions and the diffusional characteristics of more general unsteady combustion problems. A two-step reaction process is used so that the effects of stiffness can be parametrically assessed. Flame thickness and speed are easily varied from case to case by changing reaction rate parameters or initial conditions, thereby allowing efficiency assessments to be made with various ratios of flame thickness to computation space. Additionally, the problem is framed in a dimensionless form with constant physical properties, which avoids many of the complexities associated with example problems found in the literature.

Perhaps the most significant weakness in the model problem as presently formulated is the absence of convective terms. These important terms are present in the more general case of combustion in a gaseous medium and may strongly influence the effectiveness of a particular numerical method. Their absence means that conclusions drawn from the analysis of this model problem must be used with caution. However, on the positive side, techniques which show good characteristics when applied to the model problem should be considered as strong candidates for problems of a more general nature.

In this paper, five solution procedures are described and applied to the model problem. Uniform spatial grids are used with all methods. The use of adaptive gridding[11,12] shows great promise, but since that work is in the early stages of development it was considered premature to include adaptive grids in these calculations. However, some implications of recent developments will be qualitatively discussed in a later section.

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For the most part, the accuracy comparisons given herein are made on the basis of flame speed. Additional comparisons of calculated temperature and concentration profiles have been made and results from two cases are shown. The relative efficiency of the various methods is established on a case-by-case basis by determining threshold values of control parameters such as time step and grid size which will yield converged solutions. The computation time required to generate the solution using these values of control parameters is then used as the measure of efficiency. All computer times shown throughout are given in terms of seconds of CDC 6600 central processor execution time.

## Model Problem

The model problem developed in this section is a simple but significant extension of a previous problem given by Dwyer and Sanders.[11] Here two chemical reactions are considered, whereas they considered only one. The inclusion of the second reaction offers the opportunity to investigate systematically the effect of stiffness. When the rates of the two reactions differ greatly, some variables will change on time scales very different from others and the governing equations will be stiff.[13]

Consider a one-dimensional slab composed entirely of material $A$. This slab will be heated from the right-hand side. As the temperature rises material $A$ will exothermically decompose by the process $A \rightarrow B \rightarrow C$. The density of the slab will remain constant throughout the process and the properties of materials $A$, $B$, and $C$ are assumed to be equal and constant at all times.

A reaction zone (flame) will proceed through the slab until all of material $A$ is converted to material $C$. The speed at which the reaction zone moves (flame speed) will generally reach a constant value which will be maintained until the flame approaches the left side of the slab. This period of constant flame speed offers a convenient opportunity for assessing the accuracy of various numerical techniques.

In dimensional form the model problem is described by the following equations:

$$\frac{\partial \hat{\rho}_A}{\partial \hat{t}} = D_A \frac{\partial^2 \hat{\rho}_A}{\partial \hat{x}^2} - \hat{\rho}_A K_{0I} e^{-E_I/R\hat{T}} \tag{1a}$$

$$\frac{\partial \hat{\rho}_B}{\partial \hat{t}} = D_B \frac{\partial^2 \hat{\rho}_B}{\partial \hat{x}^2} + \hat{\rho}_A K_{0I} e^{-E_I/R\hat{T}} - \hat{\rho}_B K_{02} e^{-E_2/R\hat{T}} \tag{1b}$$

$$C_p \rho_0 \frac{\partial \hat{T}}{\partial \hat{t}} = k \frac{\partial^2 \hat{T}}{\partial \hat{x}^2} + \Delta h_I^0 \hat{\rho}_A K_{0I} e^{-E_I/R\hat{T}}$$

$$+ \Delta h_2^0 \hat{\rho}_B K_{02} e^{-E_2/R\hat{T}} \tag{1c}$$

where $\hat{\rho}_A$ and $\hat{\rho}_B$ are the densities of $A$ and $B$, respectively; $\hat{T}$ is temperature $\hat{t}$ and $\hat{x}$ are the temporal and spatial independent variables; $\Delta h^0$ is the heat of combustion; $D_A$ and $D_B$ are the mass diffusivities of $A$ and $B$; $k$, $C_p$, and $\rho_0$ are the thermal conductivity, specific heat, and density of the material; and $E_I$ and $E_2$ are the activation energies.

It is further assumed that the diffusion coefficients are the same for all species and that the Lewis number is unity ($\alpha/D = 1$). The following dimensionless variables are introduced:

$$\rho_A = \frac{\hat{\rho}_A}{\rho_0}, \quad \rho_B = \frac{\hat{\rho}_B}{\rho_0}, \quad X = \frac{\hat{x}}{L}, \quad \alpha = \frac{k}{\rho_0 C_p}, \quad t = \frac{\hat{t}\alpha}{L^2}$$

$$T = \frac{\hat{T} C_p}{\Delta h_I^0 + \Delta h_2^0}, \quad \theta_I = \frac{E_I C_p}{R(\Delta h_I^0 + \Delta h_2^0)}, \quad \theta_2 = \frac{E_2 C_p}{R(\Delta h_I^0 + \Delta h_2^0)}$$

$$\beta = \frac{\Delta h_I^0}{\Delta h_2^0}, \quad A_I = \frac{K_{0I} L^2}{\alpha}, \quad A_2 = \frac{K_{02} L^2}{\alpha}$$

Equations (1) can now be written in the following dimensionless form:

$$\frac{\partial \rho_A}{\partial t} = \frac{\partial^2 \rho_A}{\partial x^2} - A_I \rho_A e^{-\theta_I/T} \tag{2a}$$

$$\frac{\partial \rho_B}{\partial t} = \frac{\partial^2 \rho_B}{\partial x^2} + A_I \rho_A e^{-\theta_I/T} - A_2 \rho_B e^{-\theta_2/T} \tag{2b}$$

$$\frac{\partial T}{\partial t} = \frac{\partial^2 T}{\partial x^2} + \left(\frac{\beta}{1+\beta}\right) A_I \rho_A e^{-\theta_I/T} + \left(\frac{1}{1+\beta}\right) A_2 \rho_B e^{-\theta_2/T} \tag{2c}$$

These equations have the convenient property that $\rho_A$ and $\rho_B$ are bounded by zero and unity, and in the absence of external heat addition $T_0 \le T \le T_f$, where $T_f = T_0 + 1$.

The boundary conditions to be used are $\partial \rho_A / \partial x = \partial \rho_B / \partial x = \partial T / \partial x = 0$ at $x = 0$ and $\partial \rho_A / \partial x = \partial \rho_B / \partial x = 0$, $T = T_0 + C_I t (t < 1/C_I)$, $T = T_f (t \ge 1/C_I)$ at $X = 1$.

The value of the dimensionless parameters used in this investigation were chosen to be representative of a hydrocarbon fuel burning in air where temperature increases of a factor of 6 are fairly typical. Therefore, in most cases $T_0 = 0.2$ was used as the initial condition which results in $T_f/T_0 = 6$ except near the right boundary. A typical dimensionless activation energy for a hydrocarbon-air reaction is found from the relation

$$\theta = EC_{p \text{ mixture}} / R \Delta h_{\text{fuel}}^0 \chi$$

where $\chi$ is the moles of fuel per mole of mixture. Using typical values

$$\theta = \frac{(25 \text{ Kcal/mole})(8 \text{cal/mole} - \text{K})}{(2 \text{cal/mole} - \text{K})(357 \text{ Kcal/mole})(0.07 \text{moles/mole})} = 4$$

Values of $\theta = 3$ and 6 are also used to test sensitivity. For the most part, reaction rates are varied by varying the preexponential constants $A_I$ and $A_2$.

Two values of $\beta$, the ratio of the heats of combustion for the two reactions, are used: 0 and 1, $\beta = 0$ corresponds to all of the heat being released by the second reaction. This is similar to the procedure used by Creighton[14] in his two-stage model of methane-air reactions. $\beta = 1$ corresponds to equal heat released by each reaction and is similar to the two-stage propane-air model of Friedman and Burke.[15]

The single reaction case of $A \rightarrow B$ is formed as a special case of Eqs. (2) by letting $A_2 = 0$ and $\beta$ be large so that $\beta/(1 + \beta) \simeq 1$. The same boundary conditions previously given for $\rho_A$ and $T$ apply directly.

Each calculation is begun by applying the linearly increasing temperature at the right-hand boundary. As heat diffuses into the slab, the rate of chemical reaction increases rapidly. The resulting heat release steepens the temperature gradient, which causes a more rapid diffusion of heat into the interior. The result is an acceleration of the reaction zone to the left. After a short period, an equilibrium condition between reaction and diffusion is attained and the reaction zone (flame) then moves to the left at a constant speed. This condition is usually established by the time the reaction reaches $x = 0.7$. The flame continues to move at this speed until the effect of the left boundary is felt. This generally does not occur before the reaction is complete at $x = 0.3$. The period of motion between $x = 0.7$ and 0.3 offers an opportunity to compare steady-state flame shape and speed as calculated by the five methods.

Typical temperature profiles observed during the ignition and steady motion periods are shown in Fig. 1. Flame speeds are determined by measuring the distance between temperature profiles. In practice, interpolation is required. It is
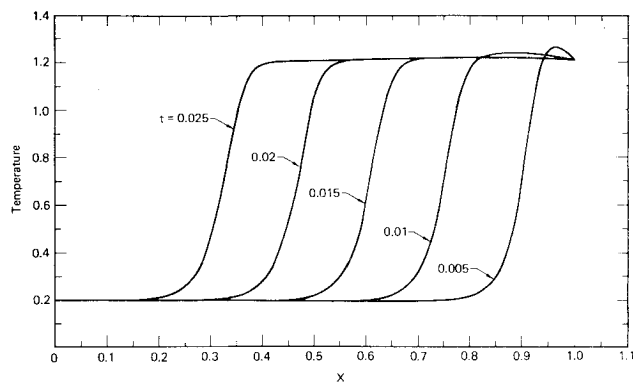
**Fig. 1 Flame propagation as indicated by temperature profiles** ($\theta_I = 4$, $A_I = 2.2 \times 10^5$).

estimated that the interpolation error in calculated flame speed is less than $\pm 1/3\%$.

## Description of Solution Procedures

The choice of numerical method is strongly influenced by the exponential nature of the reaction and the steep temperature and concentration gradients because of the slowness of diffusion compared to reaction rates. Formulating the solution technique so that a sophisticated ordinary differential equation solver can be used to integrate the reaction terms seems to offer a promising approach. This will be a basic ingredient in four of the five methods. In the fifth, the partial differential equations are replaced by the linearized difference equations which are solved as a block tridiagonal system.

The structure of the temperature profile and species concentrations within the reaction zone plays a critical role in determining flame speed even for the single reaction case. Therefore, spatial approximation and grid size are just as important in the modeling process as the treatment of the temporal portion of the equations. In Ref. 16 the results of testing both second- and fourth-order correct spatial approximations with the various methods are reported in detail. In this paper only the results of fourth-order approximations will be discussed. For simplicity, the mathematical development of the five solution procedures will be illustrated using the model problem with a single chemical reaction. Modifications required to include the second reaction will be discussed later.

### Method of Lines with Central Differences

The basic notion in the method of lines is to replace the spatial derivatives with an appropriate approximation which relates the dependent variables at neighboring grid points, then solve the resulting time-dependent ordinary differential equations as a coupled set. Consider the following differencing scheme

$$\frac{d\rho_A}{dt} = \frac{1}{12\Delta x^2} \delta_x^4 \rho A_i^n - A_I \rho_A e^{-\theta_I/RT} \tag{3a}$$

$$\frac{dT}{dt} = \frac{1}{12\Delta x^2} \delta_x^4 T_i^n + A_I \rho_A e^{-\theta_I/RT} \tag{3b}$$

where

$$\delta_x^4 \phi_i^n = -\phi_{i-2}^n + 16\phi_{i-1}^n - 30\phi_i^n + 16\phi_{i+1}^n - \phi_{i+2}^n$$

These equations can be written at each of the $N$ grid points. This procedure reduces the partial differential equations to an initial-value system of $2N$ coupled ordinary differential equations which may be solved using any appropriate in-

tegrator. In this work the ODE code (a program for solving nonstiff systems developed by Shampine and Gordon[17]) is used for cases involving only a single chemical reaction. For cases involving two-step reactions where the effects of stiffness are evaluated, a code developed by Hindmarsh[18] based on the techniques of Gear[19] is used.

The approach just described will be referred to throughout this paper as the MOL code. This procedure has the advantage of retaining the sophisticated time step and error control of the highly developed ODE and Hindmarsh-Gear programs. The obvious disadvantage is that the resulting set of coupled equations is usually very large.

### Method of Lines with Splines

A method of lines technique with a more sophisticated spatial approximation can be obtained using splines (piecewise polynomials) instead of finite-difference analogs. Madsen and Sincovec have made use of such a procedure in their development of a code known as PDECOL.[20] This code is based on the method of lines and uses a finite-element collocation procedure for discretization of the spatial variable. The assumption is made that each dependent variable may be represented by a piecewise polynomial and can be written as

$$u(x,t) = \sum_{i=1}^{NCPTS} C_i(t) \psi_i(x) \tag{4}$$

where

NCPTS = (KORD) (NINT) – (NCC) (NINT – 1)
NORD = order of the polynomials
NINT = number of intervals in the spatial domain
NCC = number of continuity conditions (e.g., NCC = 2 requires that the function and its first derivative be continuous at each grid point)
$\psi_i(x)$ = spline basis functions

The semidiscrete equations which determine the coefficients $C_i(t)$ are obtained by requiring that $u$ satisfy the partial differential equations exactly at each grid point. This results in a set of ordinary differential equations with $C_i(t)$ as the dependent variables. Once these coefficients are known, Eq. (4) can be solved since $\psi_i(x)$ are the known basis functions. The advantage of this procedure is that of a better spatial approximation, hence fewer grid points are required.

In this investigation the PDECOL code has been used without modification. Fourth-order polynomials and two continuity conditions are used for all of the cases shown. This results in fourth-order correct spatial approximation because of a special choice of collocation points within each subinterval (Gauss-Legendre quadrature points).

PDECOL calls the Hindmarsh-Gear code to integrate the system of ordinary differential equations for $C_i(t)$. Therefore, the time step and error control tests incorporated in the Hindmarsh-Gear code apply to the time discretization error. There is no error control within PDECOL of spatial discretization error. The magnitude of these errors is determined by the choice of the number of grid points, the number of continuity conditions, and the order of the polynomials.

The PDECOL code has been applied in a time-dependent approach to a steady, flat flame problem by Margolis.[21] Bledjian[22] solved the same problem using a method similar to the MOL procedure but with second-order correct differences and a Runge-Kutta integrator. Flame speeds calculated by these investigators differed by 10%.

### Operator Splitting

The operator splitting (fractional step) difference method has been applied to the modeling of premixed, laminar flame

propagation by Dwyer and Sanders,[7] and to steady, laminar, diffusion flames by Kee and Miller.[23] Similar splitting techniques have been developed by Rizzi and Bailey[24] for inviscid reacting flows and by Thomas and Wilson[25] for chemically reacting turbulent jets. The theoretical basis for the basic method has been carefully documented by Yanenko.[26]

The method used here (called majorant splitting by Yanenkó) allows one or more terms to be considered alone during each fractional step while the remaining terms are ignored. To illustrate the method, consider splitting the governing equations in the following manner:

$$\frac{\partial \rho_A}{\partial t} = \frac{\partial^2 \rho_A}{\partial x^2} \qquad (5a)$$

$$\frac{\partial T}{\partial t} = \frac{\partial^2 T}{\partial x^2} \qquad (5b)$$

$$\frac{d\rho_A}{dt} = -\rho_A A_I e^{-\theta_I/T} \qquad (6a)$$

$$\frac{dT}{dt} = \rho_A A_I e^{-\theta_I/T} \qquad (6b)$$

Equations (5), when differenced with a Crank-Nicholson scheme, yield two uncoupled sets of finite-difference equations. Equations (6) may be integrated at each grid point using the ODE or Hindmarsh-Gear codes. The first procedure is denoted as a diffusion operator $L_D$ and the second as a chemistry operator $L_C$. The computation can be carried out by first integrating Eqs. (6) over a time interval $\Delta t = t^{n+1} - t^n$. During this process the time step used in the integration is varied as necessary to maintain the specified accuracy and is generally much smaller than $\Delta t$. The values obtained at each grid point are denoted $\rho_{Ai}^{n+1}$ and $T_i^{n+1}$. The second fractional step is then executed by implicitly solving the finite-difference equations corresponding to Eqs. (5). Here, the fourth-order correct Padé difference analog[27]

$$\frac{\partial^2 \phi_i}{\partial x^2} = \frac{1}{\Delta x^2} \frac{\delta x^2}{1 + \delta x^2/12} \phi_i$$

is used for the spatial approximation where

$$\delta x^2 \phi_i = \phi_{i+1} - 2\phi_i + \phi_{i-1}$$

The resulting difference equations take the following form

$$\left(-\gamma + \frac{1}{12}\right)\phi_{i-1}^{n+1} + \left(2\gamma + \frac{5}{6}\right)\phi_i^{n+1} + \left(-\gamma + \frac{1}{12}\right)\phi_{i+1}^{n+1}$$

$$= \left(\gamma + \frac{1}{12}\right)(\phi_{\overline{i-1}}^{n+1} - 2\phi_i^{n+1} \phi_{\overline{i+1}}^{n+1}) + \phi_i^{\overline{n+1}} \qquad (7)$$

where $\phi$ corresponds to $\rho_A$ or $T$ in Eqs. (5a) and (5b), respectively, and $\gamma = \Delta t/2\Delta x^2$. The two uncoupled sets of tridiagonal difference equations can be solved for the values of the dependent variables at the $n+1$ time level. Thus, it can be seen that the solution has been advanced one time step by the sequence

$$u^{n+1} = L_D L_C u^n \qquad (8)$$

The operator splitting method retains the advantage of integrating the reaction terms with a sophisticated ordinary differential equation solver. Moreover, since the set of reaction terms is integrated separately at each grid point, the difficulty of handling large sets of ordinary differential

equations is avoided. Another advantage is that the diffusion operator has the stability normally associated with implicit methods. However, the accuracy of the method will depend on both time step and grid size. It can be seen intuitively that large time steps will tend to result in excessive uncoupling of the diffusion and chemistry processes. This can lead to significant error and eventually to complete instability.

### The Explicit/ODE Method

The explicit/ODE method is a very simple technique formulated by evaluating the spatial derivatives at the $n$ level in time, then integrating the resulting ordinary differential equations using the ODE or Hindmarsh-Gear code at each grid point over the interval $t^n$ to $t^{n+1}$. For example, the equations can be written at each grid point as

$$\left(\frac{d\rho_A}{dt}\right)_i = \left(\frac{\partial^2 \rho_A}{\partial x^2}\right)_i^n - (A_I \rho_A e^{-\theta_I/T})_i \qquad (9a)$$

$$\left(\frac{dT}{dt}\right)_i = \left(\frac{\partial^2 T}{\partial x^2}\right)_i^n + (A_I \rho_A e^{-\theta_I/T})_i \qquad (9b)$$

The spatial derivatives may be evaluated by an appropriate method. In this work, fourth-order correct central differences are used. The values thus obtained are assumed to remain constant during integration of Eqs. (9) over the interval $\Delta t$. The spatial derivatives are then re-evaluated at the $n+1$ time level, and the process is repeated.

This scheme has the advantage of simplicity and also avoids the problems associated with evaluation of large sets of ordinary differential equations. Unlike the operator splitting method, it allows diffusion to take place during the integration of the reaction terms. However, it suffers in that the integration process must be interrupted to re-evaluate the spatial derivatives frequently enough to satisfy a stability limitation imposed by the diffusion terms. If extended to problems involving combustion in a gaseous medium, the Courant-Friedrich-Lewy condition also would apply and in most cases would be the limiting restriction on $\Delta t$.

### The Linear Block Method

This method has been successfully applied to the compressible, Navier-Stokes equations by Beam and Warming[28-30] and to the equations for three-dimensional, supersonic flow by McDonald and Briley.[31] The two governing equations may be written in the form

$$\frac{\partial u}{\partial t} = \frac{\partial^2 u}{\partial x^2} + F(u)$$

where

$$u = \begin{bmatrix} P_A \\ T \end{bmatrix} \text{ and } F = \rho_A A_I e^{-\theta_I/T} \begin{bmatrix} -1 \\ 1 \end{bmatrix}$$

A second-order correct, temporal finite-difference equation is formulated using Crank-Nicholson differencing as

$$\frac{\Delta u^n}{\Delta t} = \frac{1}{2} \frac{\partial^2}{\partial x^2} (u^{n+1} + u^n) + \frac{1}{2} (F^{n+1} + F^n)$$

where $\Delta u^n \equiv u^{n+1} - u^n$. The last term is linearized by

$$F^{n+1} = F^n + \left(\frac{\partial F}{\partial u}\right)^n \Delta u^n + O[\Delta t^2]$$

Replacing the spatial derivatives with fourth-order correct, Padé difference analogs yields a coupled set of $2 \times 2$ block tridiagonal difference equations. This set may be conveniently solved by use of the bi-tridiagonal algorithm given by von

Rosenberg.[32] The Jacobian matrix is given by

$$\left(\frac{\partial F}{\partial u}\right)_i^n = A_I e^{-\theta_I/T_i^n} \begin{bmatrix} -1 & -\rho_A \theta_I/T^2 \\ 1 & \rho_A \theta_I/T^2 \end{bmatrix}_i^n$$

and the values of the dependent variables at the new time level are simply $u^{n+1} = u^n + \Delta u^n$.

The linear block method should have the increased stability associated with implicit methods and the advantage that all of the terms are coupled during the solution process. However, the linearization of the reaction terms is certain to degrade accuracy as larger time steps are taken.

## Results for Single Reaction Cases

Flame speeds as calculated by the five methods are compared in Table 1 for a wide range of reaction rates. Converged values of flame speed for each method are defined as those for which no change is observed in the first three significant figures when grid size is reduced by 20%, time step is halved, and integrator error control is tightened by an order of magnitude. Typical calculated temperature profiles are shown in Fig. 2. Similar agreement has been obtained for the other reaction rates investigated whenever the comparison is made on the basis of converged flame speed.[16] Thus, it has been concluded that all of the methods are adequate in predicting flame speed and shape.

To compare efficiency, threshold values of control parameters such as time step, number of grid points, integrator error control, etc., must be determined by trial and error. In this operation, the time step is varied in increments of a factor of 2, grid size by about 20%, and integrator error control by an order of magnitude until the most efficient combination which yields agreement within 1% of the converged value of flame speed is obtained. The results of several hundred trials of this nature are given in Ref. 16. (The relative efficiency of the five methods is about the same regardless of whether the accuracy requirement on flame speed is specified as 1% or 5%.) The results of such a procedure are shown in Table 2. To gain a clearer comparison of relative efficiency computation time normalized to that required by the linear block method is shown in Table 3. Also shown is the steady-state flame thickness $\delta_f$ corresponding to each reaction rate. (The value of $\delta_f$ used in this study is the distance between the intersections of the steepest tangent to the temperature profile and the lines defining $T_0$ and $T_f$.) Because of the normalization $\delta_f$ is representative of the ratio of reaction zone thickness to computation space.
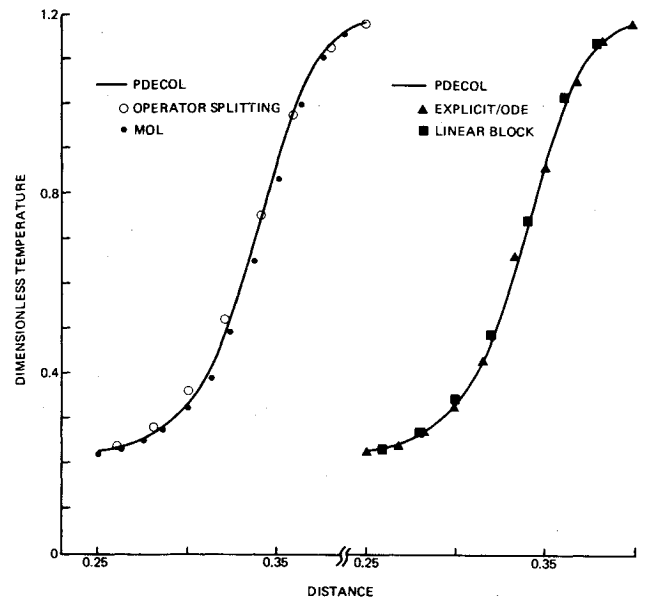


Fig. 2 Comparison of temperature profiles calculated by the five methods.

From Table 3 it can be seen that the linear block method is by far the most efficient at lower reaction rates; however, this advantage decreases substantially as $\delta_f$ becomes small. The relative increase in efficiency of the pointwise integration methods (explicit/ODE and operator splitting) is primarily because the integration need not be performed at those points ahead of the reaction zone, whereas the linear block solution involves all points. (In this problem no significant chemical reaction is taking place ahead of the reaction zone; therefore, the chemical operator need not be applied in this region. This may not be the case in more comprehensive combustion problems. If integration ahead of the reaction zone is required, the pointwise methods would need about twice as much computer time as shown.[16]) The method of lines codes also appear to benefit from the fact that little change is occurring at most grid points when the reaction zone is thin compared to the computation space.

## Results of Two Reaction Cases

The modification to the previously described solution procedures required to accommodate the second chemical

Table 1  Converged values of flame speed for five reaction rates ($T_0 = 0.2$, $\theta_I = 4$)

| Method | $A_I =$ | $2.2 \times 10^5$ | $8.8 \times 10^5$ | $19.8 \times 10^5$ | $35.2 \times 10^5$ | $400 \times 10^5$ |
|---|---|---|---|---|---|---|
| PDECOL | | 35.5 | 70.6 | 105.5 | 141.0 | ... |
| MOL | | 35.4 | 70.8 | 106.1 | 140.6 | 474 |
| Explicit/ODE | | 35.4 | 70.7 | 106.0 | 140.7 | 477 |
| Operator splitting | | 35.3 | 70.7 | 106.3 | 141.4 | 475 |
| Linear block | | 35.3 | 70.5 | 106.0 | 141.1 | 476 |

Table 2  Comparison of computation times based on conditions necessary to obtain converged values of flame speed ($T_0 = 0.2$, $\theta_I = 4$)

| Method | $A_I = 2.2 \times 10^5$ | | | $A_I = 8.8 \times 10^5$ | | | $A_I = 19.8 \times 10^5$ | | | $A_I = 35.2 \times 10^5$ | | | $A_I = 400 \times 10^5$ | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | $N$ | $\Delta t$ | $CT^a$ | $N$ | $\Delta t$ | $CT$ | $N$ | $\Delta t$ | $CT$ | $N$ | $\Delta t$ | $CT$ | $N$ | $\Delta t$ | $CT$ |
| PDECOL | 41 | ... | 25 | 61 | ... | 55 | 81 | ... | 115 | 81 | ... | 160 | ... | ... | ... |
| MOL | 81 | ... | 15 | 101 | ... | 17 | 161 | ... | 46 | 181 | ... | 55 | 600 | ... | 670 |
| Explicit/ODE | 51 | $1 \times 10^{-4}$ | 13 | 81 | $5 \times 10^{-5}$ | 14 | 121 | $1.5 \times 10^{-5}$ | 38 | 161 | $1 \times 10^{-5}$ | 49 | 500 | $1 \times 10^{-6}$ | 530 |
| Operator splitting | 51 | $4 \times 10^{-4}$ | 6.5 | 101 | $8 \times 10^{-5}$ | 14 | 161 | $4 \times 10^{-5}$ | 29 | 181 | $2 \times 10^{-5}$ | 39 | 600 | $2 \times 10^{-6}$ | 460 |
| Linear block | 51 | $2 \times 10^{-4}$ | 2.1 | 101 | $4 \times 10^{-5}$ | 8.3 | 141 | $2 \times 10^{-5}$ | 17 | 181 | $1 \times 10^{-5}$ | 33 | 600 | $1 \times 10^{-6}$ | 360 |

$^a CT \sim$ CDC 6600 central processor execution time in seconds.

**Table 3 Comparison of normalized computation time**
$(T_0 = 0.2, \theta_1 = 4)$

| | Normalized computation time | | | | |
|---|---|---|---|---|---|
| $A_1 \times 10^{-5}$ | 2.2 | 8.8 | 19.8 | 35.2 | 400 |
| $\delta_f$ | 0.070 | 0.035 | 0.026 | 0.018 | 0.005 |
| PDECOL | 12.0 | 6.6 | 6.5 | 4.8 | ... |
| MOL | 7.1 | 2.1 | 2.7 | 1.7 | 1.9 |
| Explicit/ODE | 6.2 | 1.7 | 2.2 | 1.5 | 1.5 |
| Operator splitting | 3.1 | 1.7 | 1.7 | 1.2 | 1.3 |
| Linear block | 1 . 0 | 1.0 | 1.0 | 1.0 | 1.0 |

**Table 4 The effect of stiffness on relative efficiency**
$(T_0 = 0.2, \theta_1 = \theta_2 = 4, \beta = 1)$

| $A_1 = 2.2 \times$ | $10^7$ | $10^5$ | $10^5$ | $10^5$ |
|---|---|---|---|---|
| $A_2 = 2.2 \times$ | $10^5$ | $10^5$ | $10^8$ | $10^{10}$ |
| $A_2/A_1$ | $10^{-2}$ | 1 | $10^3$ | $10^5$ |
| $U_f$ | 105.3 | 27.8 | 35.3 | 35.4 |
| $\delta_f$ | 0.033 | 0.084 | 0.068 | 0.068 |
| Computer time for linear block method (CDC 6600, s) | 89 | 4.2 | 8.2 | 8.2 |

| Method | Normalized computer time | | | |
|---|---|---|---|---|
| PDECOL | 2.5 | 7.1 | 13.0 | 11.0 |
| MOL | 3.1 | 4.3 | 7.2 | 6.3 |
| Explicit/ODE | 3.0 | 8.3 | 6.2 | 5.9 |
| Operator splitting | 2.2 | 7.7 | 5.6 | 6.1 |
| Linear block | 1.0 | 1.0 | 1.0 | 1.0 |



Fig. 3 Calculated values of trace species.

reaction are rather minor. For those methods requiring the integration of ordinary differential equations, the ODE code is replaced by the Hindmarsh-Gear code since stiff systems are to be evaluated. The increase in computation time incurred is about a factor of 2 for nonstiff cases, but great savings are realized when the system becomes stiff. In the MOL and PDECOL procedures a special feature of the Hindmarsh-Gear code is used where only a banded matrix is solved to determine the Jacobian. The finite-difference equations for the operator splitting method remain uncoupled and may be solved individually. The difference equations for the linear block method are, however, coupled. They are solved simultaneously using the tri-tridiagonal algorithm given by von Rosenberg.[32]

To gain an estimate of the effect of stiffness on the relative efficiency of the five methods, a series of cases has been analyzed where the ratio of the rates of the two reactions is varied over seven orders of magnitude. Typical results are shown in Table 4 where it can be seen that the linear block method is the most efficient regardless of which reaction is faster. For cases where $A_2 \geq A_1$, the relative advantage of the linear block method appears much the same as that observed for the slower single reaction case shown in Table 3. Notice that for these cases the thickness of the reaction zone compared to the computation space as indicated by $\delta_f$ is roughly the same.

When $A_1 \gg A_2$, faster speed and thinner flames are obtained. From examination of Table 3 it appears that stiffness has less effect on the relative efficiency of the five methods than the thickness of the reaction zone compared to the computation space. This conclusion has been supported by the other cases examined in this study.[16]

Cases where $A_2 \gg A_1$ offer an opportunity to compare the accuracy of the methods in predicting the trace species. For example, when $A_2/A_1 = 10^5$, the second reaction is so fast that species $B$ is present in amounts on the order of parts per million (ppm). Figure 3 compares values of species $B$ across the flame as calculated by the five methods. Here $\rho_B$ is plotted
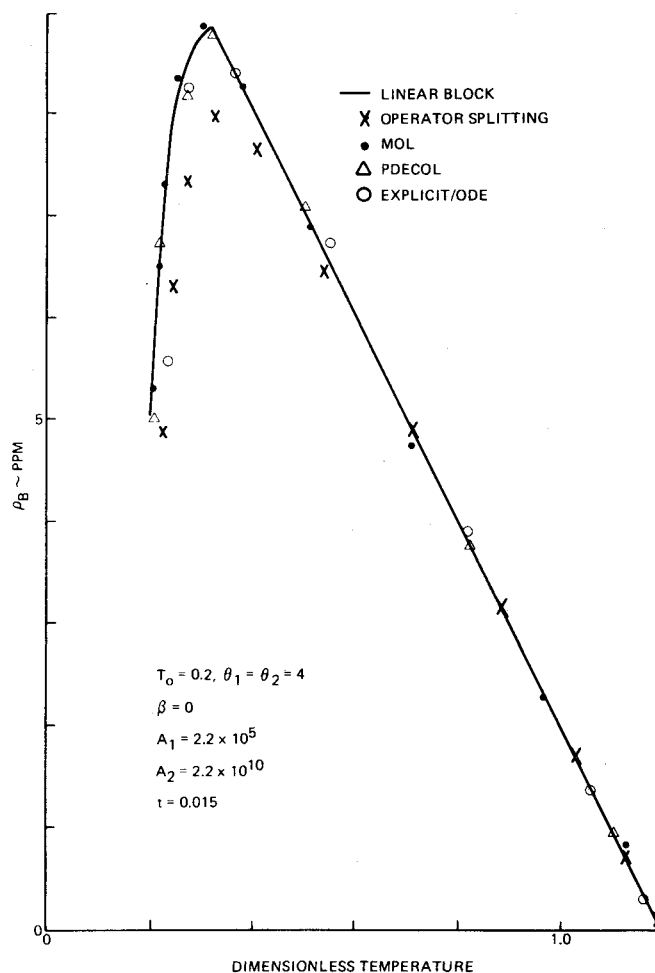
as a function of temperature to remove the effect of small variations in flame speed. All of the methods appear to do a good job of predicting trace species. Operator splitting, the only method which does not solve the equations of diffusion and reaction simultaneously, gives results which differ slightly (10% at most) from those obtained by the other four procedures.

## Discussion of Results

The results of this study indicate that the linear block method is the most efficient of the five investigated for solving the model problem. It is clearly superior for all two reaction cases investigated and for those single reaction cases where $\delta_f > 0.05$. For single reaction cases where $\delta_f < 0.05$, the MOL, explicit/ODE, and operator splitting methods become competitive.

For all of the single reaction cases and for those two reaction cases where the second reaction rate is equal to or greater than the first, it has been found for the linear block method that $\Delta X = \delta_f/5$ always provides enough grid points for spatial convergence. For certain cases where the rate of the first reaction is much greater than the second, approximately 25% more grid points may be needed. The number of grid points required for the MOL, explicit/ODE, and operator splitting methods is somewhat similar. The PDECOL code requires the fewest number of grid points; but, in almost every case investigated, it was the least efficient method.

Considering that the linear block method is superior when $\delta_f > 0.05$ and that $\Delta X \leq \delta_f/5$, then it can be concluded that the linear block method is clearly preferred when the number of grid points $N$ for the spatial coordinate is $\leq 100$. With present computing capabilities $N = 100$ in each coordinate direction is

about a practical upper limit for two-dimensional flame propagation problems. Thus, the linear block method should be considered a prime candidate for such problems provided that the extension does not introduce new complexities. Fortunately, the technique of approximate factorization[28-30] makes the extension appear rather straightforward and the advantages shown in solving the model problem can be expected to prevail when an additional spatial coordinate is introduced.

At this point it might be asked if a grid of $100 \times 100$ is adequate for simulating combustion in, say, a typical internal combustion engine. A chamber radius of 0.05 m is fairly typical and turbulent flame thicknesses of the order of $10^{-3}$ m might be expected. Using the previous estimate for $\Delta X$ yields $N = 250$. Some additional scheme for reducing the number of grid points is clearly needed. Recent developments with adaptive grids appear promising. Dwyer and Sanders[11] have presented a scheme which uses the temperature gradient in the solution to concentrate the points in the reaction zone where the temperature is changing rapidly. They report that their adaptive scheme gives results comparable with uniform methods containing three to five times as many grid points. Thus, using the linear block method with adaptive gridding appears to be a promising approach in both one- and two-dimensional models.

The effect of the convective terms on the efficiency of the linear block method when applied to problems of combustion in a gaseous medium must be assessed using a more comprehensive problem. However, some observations can be made. Convective terms introduce two types of restrictions: 1) the Courant-Friedich-Lewy condition, $\hat{c}\Delta \hat{t}/\Delta \hat{X} \leq 1$ and 2) a cell Reynolds number condition, $\hat{U}\Delta \hat{X}/\nu \leq 2$. The time step used in the solution to the model problem was of the order of that corresponding to the diffusion condition $\alpha \Delta \hat{t}/\Delta \hat{X}^2 \leq \frac{1}{2}$. For most gaseous combustion processes, the time step limitation imposed by the Courant condition is smaller than that given by the diffusion condition (for $N = 100$). Butler and O'Rourke[9] report taking time steps 16 times the Courant number using their implicit formulation on a one-dimensional flame propagation problem. However, even steps this large are usually smaller than those found necessary to solve the reaction-diffusion terms alone. The Courant restrictions should apply equally to the linear block and operator splitting methods—a significant disadvantage for operator splitting, since in solving the model problem, its time step size was usually twice that used for the linear block method. The Courant condition places a definite limitation on the explicit/ODE method. If the implicit methods can use a time step considerably larger than that implied by the Courant condition, this method would no longer be competitive.

Whether a cell Reynolds number problem exists can be estimated in the following way. Assuming for a moment that the fluid velocity is equal to the flame speed, the restriction may be written

$$\hat{U}_f \Delta \hat{X}/\nu \leq 2$$

Simple flame theory[33] predicts that $\hat{U}_f \hat{\delta}_f \approx \alpha$; and, as previously discussed, $\Delta \hat{X} \approx \hat{\delta}_f/5$; therefore, no difficulty is encountered unless $\alpha/\nu > 10$. But $\nu/\alpha$ is the Prandtl number and for applications of interest is of order unity. Therefore, the grid size previously determined by the reaction-diffusion model need not be reduced because of cell Reynolds number restriction unless the fluid velocity is approximately 10 times the flame speed in a particular coordinate direction.

## Concluding Remarks

A reaction-diffusion model problem has been used to evaluate the relative efficiency and accuracy of five numerical solution procedures for modeling flame propagation. Good agreement in calculated values of flame speed and temperature and concentration profiles has been obtained for all of the methods, but the linear block method was found to be significantly more efficient in terms of computer time over a wide range of reaction rates for both global and two-step chemical kinetics.

The linear block method appears to be compatible with adaptive gridding procedures under development. Gridding procedures of this nature may be essential for reducing the size of two-dimensional problems to a level acceptable for solution with presently available computers.

It has been surmised that the relative efficiency of the linear block method will be retained when extended to combustion processes in a gaseous medium. It also appears that the method may be readily extended to two-dimensional problems by the use of the approximate factorization technique which retains the formal accuracy of the solution process. However, validity of these conjectures must be established by applying the method to more comprehensive test problems.

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