

Application of Semi-implicit Runge–Kutta Methods for Integration of Ordinary and Partial Differential Equations

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Abstract

A computer algorithm for the integration of sets of stiff ordinary differential equations has been developed. The algorithm utilizes a semi-implicit Runge–Kutta method for the single step integrations. This algorithm in combination with discretization by orthogonal collocation is particularly well suited for the integration of non-linear parabolic partial differential equations.

1. INTRODUCTION

A variety of models for chemical engineering systems leads to stiff ordinary differential equations, *i.e.* differential equations with a large spread in the magnitude of the local eigenvalues. Kinetic systems involving extremely rapid reactions are a typical example [1] and transient models for fluid bed reactors [2] and distillation columns [3] exhibit a similar behavior. Furthermore, stiffness is encountered in the sets of ordinary differential equations that are obtained by discretization of partial differential equations. Such equations arise from, for example, two-dimensional reactor models [4, 5], transients for catalyst particles [6, 7] and a variety of problems in fixed bed adsorption [8].

Explicit integration methods such as the classical fourth order Runge–Kutta method are unsuitable for the numerical integration of such systems as they would require an often prohibitively large number of steps. A simple semi-implicit Runge–Kutta method, however, has proved to be highly effective [9, 10], and the application to various problems of an algorithm based on this technique is described in this paper.

2. THE SEMI-IMPLICIT RUNGE–KUTTA METHOD

Let the system to be integrated be

$$\frac{d}{dt} \mathbf{y} = \mathbf{f}(\mathbf{y}) \quad (1)$$

with initial element

$$\mathbf{y}(t_n) = \mathbf{y}_n \quad (2)$$

and let the Jacobian matrix \mathbf{A} be given by

$$A_{ij} = \left(\frac{\partial f_i}{\partial y_j} \right)_{\mathbf{y}_n} \quad (3)$$

The solution $\mathbf{y}_{n+1} = \mathbf{y}(t_n + h)$ is found from

$$\mathbf{k}_1 = h(\mathbf{I} - ha\mathbf{A})^{-1} \mathbf{f}(\mathbf{y}_n) \quad (4)$$

$$\mathbf{k}_2 = h(\mathbf{I} - ha\mathbf{A})^{-1} \mathbf{f}(\mathbf{y}_n + b_2\mathbf{k}_1) \quad (5)$$

$$\mathbf{k}_3 = (\mathbf{I} - ha\mathbf{A})^{-1} (b_{31}\mathbf{k}_1 + b_{32}\mathbf{k}_2) \quad (6)$$

$$\mathbf{y}_{n+1} = \mathbf{y}_n + R_1\mathbf{k}_1 + R_2\mathbf{k}_2 + R_3\mathbf{k}_3 \quad (7)$$

where the constants a , b_2 , b_{31} , b_{32} , R_1 , R_2 and R_3 are given in ref. 9.

The method was originally suggested in ref. 11. It is third order and L -stable, and each step requires one triangular decomposition and two function evaluations.

Step length adjustment is essential for an effective implementation. The full step–half step technique as described in ref. 9 is used. A complete description of the algorithm with commented subprograms is found in ref. 10.

3. APPLICATION TO ORDINARY DIFFERENTIAL EQUATIONS

The following kinetic example is based on a set of rate equations from Gelinas [1], who investigated a model for smog formation. His model originally involved 29 species and

about 60 reactions with rate constants varying by 20 orders of magnitude. The complete model was effortlessly integrated by the present algorithm but for demonstration purposes only a condensed version involving 12 components and 22 reactions is shown.

The components of the reaction are given in Table 1, and the rate constants for the first- and second-order (irreversible) reactions in Table 2.

TABLE 1

Components of the smog model

1, NO ₂	2, NO	3, O
4, O ₃	5, C ₄ H ₈	6, C ₃ H ₇ O ₂
7, HO ₂	8, CH ₃ CO ₃	9, CH ₃ O ₂
10, HO	11, C ₄ H ₈ OHO ₂	12, CH ₂ OHO ₂

TABLE 2

Reactions and rate constants

1	→	2 + 3	6.7×10^{-3}
3	→	4	6.73×10^4
2 + 4	→	1	9.1×10^{-2}
3 + 5	→	6 + 7	3.8×10^2
6 + 2	→	1 + 7	3.0
4 + 5	→	7 + 8	4.9×10^{-5}
4 + 5	→	4	2.1×10^{-5}
2 + 8	→	1 + 9	3.0
1 + 8	→	inactive	0.1
5 + 8	→	9	1.0×10^{-4}
2 + 9	→	1 + 7	3.0
5 + 10	→	11	1.0×10^2
2 + 11	→	1 + 12	3.0
2 + 12	→	1 + 10	3.0
2 + 7	→	1 + 10	50
7 + 7	→	inactive	50
7 + 10	→	inactive	50
7 + 6	→	inactive	50
7 + 9	→	inactive	50
7 + 8	→	inactive	50
7 + 11	→	inactive	50
7 + 12	→	inactive	50

Initially only components 1 and 5 are present, in concentrations of 4.0×10^{-8} and 11.2×10^{-8} mol l⁻¹ respectively. The integration proceeds smoothly to a final time $t = 3000$ s, and a computation time of about 1.5 s on the IBM 370/165 is required. Integration results for the main components y_1 , y_2 , y_4 and y_5 are given in Table 3. A complete program listing and a comparison with the full model is given in ref. 10.

TABLE 3

Integration of the kinetic problem

Time	y_1	y_2	y_4	y_5
0	4.0	0.0	0.0	11.2
1	3.97	0.027	0.03	11.2
10	3.79	0.212	0.23	11.17
100	3.81	0.177	1.08	10.57
1000	3.53	0.077	2.97	6.75
3000	2.81	0.049	3.96	2.39

Stability requirements for integration using a conventional fourth-order Runge-Kutta method limit the step size to less than about 0.0001, and the computation time would increase by at least four orders of magnitude.

An alternate stiff integrator, Gear's method [12], yields identical results using about the same computation time. Triangular decomposition is not required at each step in Gear's method, where the correction vectors are determined iteratively, and hence this method may be more economical for very large systems of equations. The semi-implicit Runge-Kutta methods, however, have the advantage of unconditional stability and of being simpler, more robust and easily adapted to special problems.

4. INTEGRATION OF PARABOLIC PARTIAL DIFFERENTIAL EQUATIONS USING ORTHOGONAL COLLOCATION

Models leading to non-linear parabolic partial differential equations are frequently encountered in chemical engineering problems. Traditionally, finite difference methods, *e.g.* the Crank-Nicholson method, have been used for their numerical solution. Such methods are easy to apply but a very fine grid is often required, resulting in excessive computation time.

Villadsen [13] and later Ferguson and Finlayson [14] proposed the efficient orthogonal collocation method as an alternative method for discretization of the spatial coordinate. Villadsen also used collocation for the integration of the resulting initial value problem, while an explicit method was used in ref. 14 and in the majority of later applications of the collocation method.

For simple problems with one dependent variable explicit methods are definitely applicable for the set of coupled ordinary differential equations obtained by low order collocation. An increase in the number of collocation points leads both to increased dimensionality and to increased stiffness of the discretized set of equations [15].

The effect of stiffness can be much more severe in problems with more than one dependent variable, since the natural time scales may differ by orders of magnitude. For such problems explicit methods may be inapplicable even using low order collocation, and compared with the semi-implicit Runge-Kutta methods integration by collocation in the time coordinate is uneconomical.

As an example we shall here consider the transient of a spherical catalyst particle where a first-order exothermic irreversible reaction occurs. This system has been investigated by Hlavacek and Marek [6] and by Lee and Luss [7], and integration using orthogonal collocation and an explicit integration method is discussed by Ferguson and Finlayson [14].

The unsteady state material and energy balances are

$$\frac{\partial y}{\partial \tau} = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial y}{\partial r} \right) - \Phi^2 y \exp \left\{ \gamma \left(1 - \frac{1}{\theta} \right) \right\} \quad (8)$$

$$\text{Le} \frac{\partial \theta}{\partial \tau} = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial \theta}{\partial r} \right) + \beta \Phi^2 y \exp \left\{ \gamma \left(1 - \frac{1}{\theta} \right) \right\} \quad (9)$$

y and θ represent the dimensionless concentration and temperature. Φ is the Thiele modulus, γ a dimensionless activation energy and β a dimensionless heat of reaction. In the present study the parameters of Lee and Luss are used: $\Phi = 1.1$, $\gamma = 30$ and $\beta = 0.15$. Surface resistances to mass and heat transfer are neglected, and the boundary conditions are

$$r = 0 \quad \frac{\partial y}{\partial r} = \frac{\partial \theta}{\partial r} = 0 \quad (10)$$

$$r = 1 \quad y = \theta = 1 \quad (11)$$

A unique steady state is obtained, but for $\text{Le} < 0.397$ the steady state is unstable and a limit cycle is found.

The initial conditions are chosen as

$$\tau = 0 \quad y = \theta = 1 \quad (12)$$

The Laplacian is discretized by orthogonal collocation [15]:

$$\begin{aligned} \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial y}{\partial r} \right) &= \sum_{j=1}^N C_{ij} y_j + C_{i,N+1} y(r=1) \\ &= \sum_{j=1}^N C_{ij} y_j + C_{i,N+1} \end{aligned} \quad (13)$$

The y_i represent the values of the dependent variable at the N collocation points, chosen as the zeros of $P_N^{0,0.5}(r^2)$ [15].

Consequently we arrive at the following set of $2N$ coupled ordinary differential equations:

$$\begin{aligned} \frac{d}{d\tau} y_i &= \sum_{j=1}^N C_{ij} y_j + C_{i,N+1} - \\ &\quad - \Phi^2 y_i \exp \left\{ \gamma \left(1 - \frac{1}{\theta_i} \right) \right\} \end{aligned} \quad (14)$$

$$\begin{aligned} \text{Le} \frac{d}{d\tau} \theta_i &= \sum_{j=1}^N C_{ij} \theta_j + C_{i,N+1} + \\ &\quad + \beta \Phi^2 y_i \exp \left\{ \gamma \left(1 - \frac{1}{\theta_i} \right) \right\} \end{aligned} \quad (15)$$

where

$$y_i(\tau = 0) = \theta_i(\tau = 0) = 1 \quad i = 1, 2, \dots, N \quad (16)$$

A similar approach was used by Ferguson and Finlayson, who obtained a numerical solution of eqns. (14) and (15) by an explicit integration method, a second-order predictor-corrector method. They concluded that collocation was far superior to a finite difference approximation.

Numerical stability of their explicit integration method requires that the step length h of the time step is chosen such that $h|\lambda_{\max}| < 2$, where λ_{\max} is the eigenvalue of largest magnitude of the Jacobian.

The value of λ_{\max} depends strongly on the collocation order N and the Lewis number Le . Wedel *et al.* [16] found that λ_{\max} is closely approximated by

$$|\lambda_{\max}| = \begin{cases} |\lambda_{c,\max}| & \text{for } \text{Le} > 1 \\ \frac{1}{\text{Le}} |\lambda_{c,\max}| & \text{for } \text{Le} < 1 \end{cases} \quad (17)$$

where $\lambda_{c,\max}$ is the numerically largest eigen-

value of the discretization matrix C for the Laplacian. $\lambda_{c,\max}$ increases rapidly with increasing N , as shown in Table 4.

TABLE 4

Numerically largest eigenvalue of the discretization matrix C

N	2	3	4	5	6	8	10
λ_{\max}	-95	-326	-835	-1784	-3386	-9538	-21700

The total number of time steps required to integrate the transient is the ratio of the response time τ_r of the system to the step length. Let us define τ_r as the time required to obtain a 99% approach to the steady state for a stable system, or the cycle time for a system exhibiting limit cycles. The number of time steps M required using $N = 5$ and for the predictor-corrector method used by Ferguson and Finlayson with the largest possible step length is shown in Table 5.

TABLE 5

Number of steps by explicit integration

Le	0.3	0.45	1	5	1000
τ_r	0.5	3.15	2.1	15	3000
λ_{\max}	-5950	-3970	-1784	-1784	-1784
M	1490	6250	1780	13400	2.7×10^6
	(per cycle)				

The large response time for $Le \gg 1$ requires a prohibitively large number of steps for an explicit method. Similarly, small values of Le would not change τ_r much but would increase the value of λ_{\max} , again resulting in a very large M . We may thus conclude that explicit methods are unfeasible for $Le \gg 1$ and for $Le \ll 1$.

The number of time steps and the computing time (IBM 370/165) for the semi-implicit method are given in Table 6. It should be noted that this method uses a variable step length which permits large steps during a smooth part of the transient. The accuracy is about 10^{-3} for $Le = 0.3$ and $Le = 0.45$ and better than 10^{-5} for $Le = 1.5$ and 1000.

Figures 1 and 2 show the average pellet concentration \bar{y} versus τ . The limit cycle at $Le = 0.3$ and the oscillatory approach at $Le =$

TABLE 6

Number of steps and integration time for the semi-implicit method

Le	0.3	0.45	1	5	1000
M	19	28	11	14	21
Computing time (s)	0.7	0.9	0.3	0.4	0.7

0.45 are the most difficult to integrate, while the fairly large M at $Le = 1000$ is caused by the widely differing time scales for the rapid concentration response and the slow temperature response. Apart from this, however, the differing time constants for the concentration and the temperature response give no problems.

Sincovec and Madsen [17] have recently developed a program system for solution of partial differential equations. They use finite difference methods for discretization and a variant of Gear's method specially suited for a banded Jacobian for the time integrations.

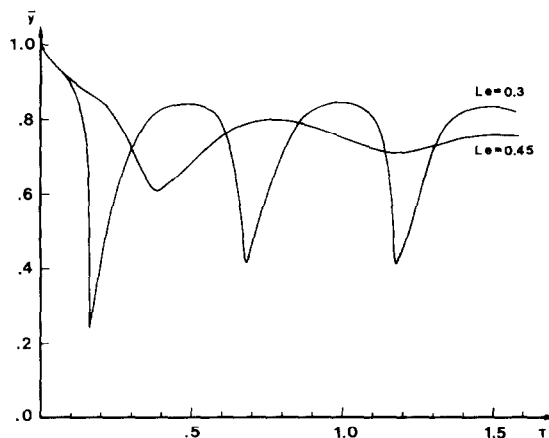


Fig. 1. Time response of the average pellet concentration for small Le .

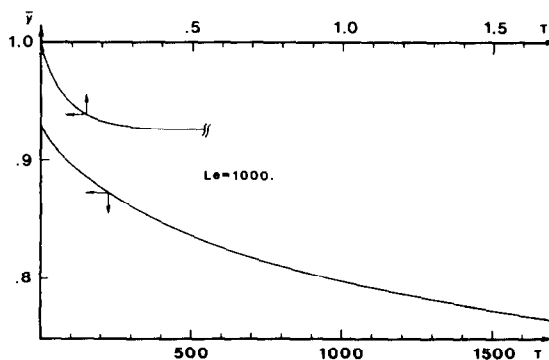


Fig. 2. Time response of the average pellet concentration for $Le = 1000$.

Discretization by collocation is probably much more efficient for the type of problems considered here, while their method may well be advantageous for problems with a large number of dependent variables.

5. OTHER PROBLEM TYPES

As a final example we shall look at a type of equation very frequently encountered in models for engineering systems.

Consider the fluid phase material balance for an isothermal fixed bed reactor with an n th order catalytic reaction:

$$\frac{dy_B}{dz} = -aR(y) \quad (18)$$

where the average rate of reaction $R(y)$ in the spherical catalyst is found from

$$0 = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial y}{\partial r} \right) - \Phi^2 y^n \quad (19)$$

with boundary conditions

$$r = 0 \quad \frac{\partial y}{\partial r} = 0 \quad (20)$$

$$r = 1 \quad y = y_B \quad (21)$$

and where

$$R(y) = \int_0^1 3r^2 y^n dr \quad (22)$$

Equation (18) is easily integrated, for example, by the classical fourth order Runge-Kutta method. However, evaluation of $R(y)$ at each step necessitates the separate solution of the non-linear differential equation (19) four times.

This problem is circumvented in the following manner. Equation (19) is converted into a pseudo "transient"

$$\epsilon \frac{\partial y}{\partial z} = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial y}{\partial r} \right) - \Phi^2 y^n \quad (23)$$

where ϵ is chosen to be very small. Discretization of eqn. (23) and evaluation of $R(y)$ by Radau quadrature now yields the following set of ordinary differential equations:

$$\epsilon \frac{dy_i}{dz} = \sum_{j=1}^N C_{ij} y_j - \Phi^2 y_i^n + C_{i,N+1} y_B \quad i = 1, 2, \dots, N \quad (24)$$

$$\frac{\partial y_B}{\partial z} = -a \left(\sum_{j=1}^N w_j y_j^n + w_{N+1} y_B^n \right) \quad (25)$$

where the y_i are initially chosen to satisfy

$$0 = \sum_{j=1}^N C_{ij} y_j - \Phi^2 y_i^n + C_{i,N+1} y_B \quad (z = 0) \quad i = 1, 2, \dots, N \quad (26)$$

Choosing, for example, $\epsilon = 10^{-5}$ yields an extremely stiff set which is nevertheless effortlessly integrated.

For the numerical example

$$\epsilon = 10^{-5}, \quad a = 5, \quad \Phi^2 = 10, \quad n = 2, \quad N = 4, \quad y_B(z = 0) = 1$$

the results given in Table 7 are found for a computing time of less than 0.1 s. The results are accurate to the number of digits shown.

TABLE 7

Fixed bed reactor with second order reaction and diffusion resistance

z	0	0.25	0.5	0.75	1.0
y_B	1	0.56939	0.38230	0.28198	0.22084

6. CONCLUSION

An integration algorithm based on a semi-implicit Runge-Kutta technique is well suited for integration of a variety of stiff problems.

In particular a combination of discretization by orthogonal collocation and integration by the semi-implicit Runge-Kutta method appears to be very attractive for numerical integration of a spectrum of chemical engineering models leading to parabolic non-linear partial differential equations.

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