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Численное моделирование химической кинетики двухстадийным методом решения неявных систем

Numerical Simulation of Chemical Kinetics With a Two-Stage Method for Solving Implicit Systems

Аннотация. В химической кинетике и в других важных приложениях возникает задача Коши для жесткой системы ОДУ неразрешенных относительно производной. Построен двухстадийный L-устойчивый метод типа Розенброка, предназначенный для решения неявных жестких систем ОДУ. На основе этого метода сформулирован алгоритм интегрирования переменного шага. Приведены результаты расчетов, подтверждающие эффективность нового алгоритма.

Ключевые слова. неявная система, метод Розенброка, контроль точности.

Abstract. In chemical kinetics and other important applications there often arises the Cauchy problem for a stiff system of ODEs unresolved with respect to derivative. The two-stage *L*-stable Rosenbrock type method is derived. On the base of the method the integration algorithm of alternating stepsize is designed. It is aimed at solving implicit stiff systems of ODEs. Numerical results confirming the efficiency of new algorithm are given.

Keywords. implicit system, Rosenbrock method, accuracy control.

Introduction

On modelling dynamic processes in chemical kinetics, electric circuits, and other areas there arises the Cauchy problem for stiff systems of differential equations unresolved with respect to derivative [1-5]:

$$F(x,x',t)=0,\ x(t_0)=x_0,\ t_0\le t\le t_k, \tag{1}$$

where x and F represent real-valued N-dimensional vector-functions, t is an independent variable. Modern numerical methods usually require the following form of the problem [1, 6-7]

$$x'=f(t,x), x(t_0)=x_0,\ \ t_0\le t\le t_k \tag{2}$$

Transforming problem (1) to form (2) usually causes additional expenses per each integration step due to decomposing matrix $F_y=\partial F(x,y,t)/\partial y$. At that, F_y often is singular. Resolved problem (2) is, as a rule, stiff.

The most known algorithms aimed at solving problem (1) are based either on implicit multistep numerical formulas [1, 2] or implicit one-step Runge-Kutta schemes [8]. In paper [9] there studied deriving non-iterative (m, k)-schemes for problem (1) solution. These schemes are similar in terms of structure to the Rosenbrock type scheme offered in this paper. Many practical problems are described by so called hybrid systems [7]. Such problems are defined by regimes which alternate each other at some points. These points are given by event functions $g(x)$ which depend on unknown solution $x(t)$. In such a situation multistep methods may be inefficient because at each regime alternation whole information about the approximate solution at previous points is lost.

Rosenbrock type methods have got widely spread on solving stiff problems. This is due to easy implementation and rather good accuracy and stability properties. It is natural to resolve problem (1) and provide L-stability of a numerical scheme at the same time to increase the efficiency of calculations. Here is derived the two-stage L-stable second order method for solving implicit problems. The difference between

new method and traditional Rosenbrock type methods is that former, in addition, numerically computes the solution derivative.

1. Rosenbrock Type Methods for Resolved Problems

For solving problem (2) Rosenbrock type methods take the form

$$x_{n+1} = x_n + \sum_{i=1}^{m} p_i k_i \,, D_n k_i = hf\left(t_n + c_i h, x_n + \sum_{j=1}^{i-1} \beta_{ij} k_j\right), \tag{3}$$

where k_i, $1 \le i \le N$, represent stages of a method, h is the integration stepsize, $D_n = I - ahf_n'$, I is the identity matrix, $f_n' = \partial f(t_n, x_n)/\partial x$ denotes the Jacobi matrix of system (2), a, c_i, p_i, β_{ij}, $1 \le i \le N$, $1 \le j \le i-1$, represent numerical coefficients defining accuracy and stability properties of (3). Nowadays the Rosenbrock type methods are treated more widely – each method involving the Jacobi matrix is related to the family of Rosenbrock type schemes or non-iterative methods [1]. For a nonautonomous problem two-stage method (3) has the form

$$\begin{aligned} &x_{n+1} = x_n + p_1 k_1 + p_2 k_2, \\ &D_n k_1 = hf(x_n), D_n k_2 = hf\left(x_n + \beta_{21} k_1\right). \end{aligned} \tag{4}$$

With the following coefficients

$$p_1 = \beta_{21} = a = 1 - 0.5\sqrt{2}\,, p_2 = 1 - a = 0.5\sqrt{2}$$

numerical schemes (4) is order 2 and, in addition, *L*-stable. For accuracy control of (4) we can apply the inequality [10]

$$\| k_2 - k_1 \| \le \varepsilon$$

where ε is the defined tolerance, $\|\cdot\|$ denotes some norm in $\mathbb{R}^n$. To evaluate increments k_i, $1 \le i \le 2$, it is necessary to solve two linear systems of algebraic equations. This is usually performed applying *LU*-decomposition.

2. Methods of the Rosenbrock type for implicit systems

Using notation $x' = y$ problem (1) can be written in the form

$$x' = y\,,\, F(x,y,t) = 0\,,\, t_0 \le t \le t_k\,. \tag{5}$$

It is the Cauchy problem for a system of DAEs with initial conditions $x(t_0) = x_0$ and $y(t_0) = y_0$. Latter condition can be obtained, for example, solving problem $F(x_0, y, t_0) = 0$ by the relaxations [10]. Below we assume the existence and uniqueness of the solution of problem (1) of (5). Also, it is assumed that function F is sufficiently differentiable at each step and matrix

$$D_n = F_{ny} + ahF_{nx}$$

is nonsingular. Here, a is a numerical coefficient, h is the integration stepsize,

$$F_{nx} = \frac{\partial F(x_n, y_n, t_n)}{\partial x},\; F_{ny} = \frac{\partial F(x_n, y_n, t_n)}{\partial y}.$$

Now, *m*-stage Rosenbrock type method applied to problem (1) or (5) takes the form [4]:

$$\begin{aligned}
&x_{n+1} = x_n + \sum_{i=1}^{m} p_i k_i^x\,,\, y_{n+1} = y_n + \sum_{i=1}^{m} p_i k_i^y\,,\\
&D_n k_i^x = hF_{ny} \cdot \left(y_n + \sum_{j=1}^{i-1} \beta_{ij} k_j^y \right) - ah^2 F_{nt} -\\
&-hF\left(x_n + \sum_{j=1}^{i-1} \beta_{ij} k_j^x\,,\, y_n + \sum_{j=1}^{i-1} \beta_{ij} k_j^y\,,\, t_n + h \cdot \sum_{j=1}^{i-1} \beta_{ij} \right),\\
&k_i^y = \frac{1}{ah}\left[k_i^x - h \cdot \left(y_n + \sum_{j=1}^{i-1} \beta_{ij} k_j^y \right) \right],
\end{aligned} \tag{6}$$

where a, p_i, and β_{ij} are numerical coefficients, $F_{nt} = \partial F(x_n, y_n, t_n)/\partial t$. Obviously, applying formulas (6) to solve (2) we get Rosenbrock type methods (3). On solving problem (5) scheme (6) numerically computes the solution derivative. Since y_0 is evaluated approximately, it is assumed that $\| F_0 \| \le C \cdot h^p$. Here $F_0 = F(x_0, y_0, t_0)$, $\|\cdot\|$ denotes some norm in $\mathbb{R}^n$, C is a constant not depending on the integration stepsize, p represents order of a method. Below we assume that F_{ny} is nonsingular over each interval $[t_n, t_{n+1}]$.

3. Two-stage Second Order Method

To solve (5) consider two-stage formula of the form

$$
\begin{aligned}
&x_{n+1} = x_n + p_1 k_1^x + p_2 k_2^x,\ y_{n+1} = y_n + p_1 k_1^y + p_2 k_2^y,\\
&D_n k_1^x = h \cdot \left[F_{ny} \cdot y_n - ahF_{nt} - F(x_n, y_n, t_n) \right],\\
&k_1^y = \frac{1}{ah} \cdot \left(k_1^x - hy_n \right),\\
&D_n k_2^x = hF_{ny} \cdot \left(y_n + \beta_{21} k_1^y \right) - ah^2 F_{nt} -\\
&-hF\left(x_n + \beta_{21} k_1^x, y_n + \beta_{21} k_1^y, t_n + \beta_{21} h \right),\\
&k_2^y = \frac{1}{ah} \cdot \left[k_2^x - h\left(y_n + \beta_{21} k_1^y \right) \right].
\end{aligned}
\tag{7}
$$

Expand stages k_1^x, k_1^y, k_2^x, and k_2^y in the Taylor series in terms of h and substitute in the first formula of (7). Performing comparison between resulting expression for approximate solution and the Taylor series for the exact solution we obtain the second order conditions for scheme (7), i.e.

$$
p_1 + p_2 = 1,\ \beta_{21} p_2 = \frac{1}{2} - a\,. \tag{8}
$$

Expanding $F_{n+1} = F\left(x_{n+1}, y_{n+1}, t_{n+1} \right)$ in the Taylor series and allowing for order conditions (8), get

$$
F_{n+1} = \frac{a^2 - 2a + 0.5}{a^2} F_n + \frac{2a + (2a-1)\beta_{21}}{4a} h^2 \varphi(t_n) + O(h^3)
$$

where function $\varphi(t)$ does not depend on the integration stepsize. The requirement $F_{n+1} = O(h^2)$ leads to relation

$$
a^2 - 2a + 0.5 = 0\,. \tag{9}
$$

The given condition provides *L*-stability of scheme (7) applied to the Dahlquist equation $x' = \lambda x$, where λ represents a complex number, $Re(\lambda) < 0$. Equation (9) has two real roots $a_1 = 1 - 0.5\sqrt{2}$ and $a_2 = 1 + 0.5\sqrt{2}$. On solving resolved problems usually former is chosen because in this case the coefficient in the main term of local

truncation error is less. It follows from numerous calculations that on solving implicit systems a_1 is also preferable.

In paper [11] it is shown that applying *L*-stable inner (intermediate) numerical schemes increases the efficiency and reliability of calculations. The intermediate scheme

$$x_{n+\beta} = x_n + \beta_{21} k_1^x$$

is *L*-stable, if $\beta_{21} = a$. Using this and allowing for order conditions (8) we derive coefficients of *L*-stable method (7) of order 2, i.e.

$$p_1 = \beta_{21} = a = 1 - 0.5\sqrt{2}\,,\ p_2 = 1 - a = 0.5\sqrt{2}\,. \tag{10}$$

4. Accuracy Control of Calculations

Accuracy control of calculations for scheme (7) can be carried out by analogy with [5], i.e. at each step it is necessary to check the following inequality

$$\| k_2^x - k_1^x \| \le \varepsilon, \tag{11}$$

where k_1^x and k_2^x have already been defined in (7), ε is the defined tolerance, norm $\| \zeta_n \|$ was computed by the formula $\| \zeta_n \| = \max\limits_{1 \le i \le N} \left(| \zeta_n^i | / (| x_n^i | + r) \right)$. If $| x_n^i | < r$, then in i-th component there controlled absolute error $r \cdot \varepsilon$, otherwise – relative error ε.

Since the solution derivative in numerical formula (7) is evaluated approximately, in addition to (11) we check the following inequality

$$\| D_n^{-1} F_n \| \le \varepsilon. \tag{12}$$

In calculations we used norm $\| \xi \|$ of the form $\| \xi_n \| = \max\limits_{1 \le i \le N} | \xi_n^i |$.

Numerical Results

The Chemical Akzo Nobel Problem was chosen to test new two-stage method that is called *iros2* below. The origin of the problem is described in [12]. The problem is given by a stiff system of 6 non-linear DAEs of index 1. It is of the form

$$M\frac{dy}{dt} = f(y),\ y(0) = y_0,\ y'(0) = y_0', \text{ with } y \in \mathbb{R}^6, 0 \le t \le 180.$$

The matrix M is of rank 5 and given by

$$M = \begin{pmatrix} 1 & 0 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \end{pmatrix}$$

and function f by

$$f_1 = -2r_1 + r_2 - r_3 - r_4,\ f_2 = -\frac{1}{2}r_1 - r_4 - \frac{1}{2}r_5 + F_{in},\ f_3 = r_1 - r_2 + r_3,$$

$$f_4 = -r_2 + r_3 - 2r_4,\ f_5 = r_2 - r_3 + r_5,\ f_6 = K_s \cdot y_1 \cdot y_4 - y_6,$$

where r_i and F_{in} are auxiliary variables given by

$$r_1 = k_1 \cdot y_1^4 \cdot \sqrt{y_2},\ r_2 = k_2 \cdot y_3 \cdot y_4,\ r_3 = \frac{k_2}{K} \cdot y_1 \cdot y_5,\ r_4 = k_3 \cdot y_1 \cdot y_4^2,\ r_5 = k_4 \cdot y_6^2 \cdot \sqrt{y_2},$$

$$F_{in} = klA \cdot \left(\frac{p(CO_2)}{H} - y_2 \right).$$

Calculations were performed using the following parameters

$$k_1 = 18.7,\ k_2 = 0.58,\ k_3 = 0.09,\ k_4 = 0.42,\ K = 34.4,\ klA = 3.3,$$

$$K_s = 115.83,\ p(CO_2) = 0.9,\ H = 737.$$

The initial vectors are

$$y_0 = (0.444, 0.00123, 0, 0.007, 0, K_s \cdot y_{0,1} \cdot y_{0,4})^T,\ y_0' = f(y_0).$$

For negative values of y_2 function f can not be evaluated and the integration stepsize is reduced until the inequality $y_2 \ge 0$ is satisfied.

According to [12] the reference solution was computed by PSIDE on a Cray C90, using double precision, $rtol = atol = 10^{-19}$. At the end of the integration interval it is of the form

$$y_1 = 0.1150794920661702,\ y_2 = 0.1203831471567715 \cdot 10^{-2},$$

$$y_3 = 0.1611562887407974,\ y_4 = 0.3656156421249283 \cdot 10^{-3},$$

$$y_5 = 0.1708010885264404 \cdot 10^{-1},\ y_6 = 0.4873531310307455 \cdot 10^{-2}.$$

Calculations were performed on Intel(R) Core(TM) i5-3317U CPU @ 1.70GHz using numerical Jacobi matrix. Parameter r in norm was set to 1 so that relative and absolute tolerances were equal. Computational costs for the Chemical Akzo Nobel Problem simulation are given in the Table 1.

Table 1. Computational costs for the Chemical Akzo Nobel Problem

Solver	**rtol**	**atol**	**scd**	**steps**	**f**	**dec**
iros2	10^{-2}	10^{-2}	2.51	27	66	33
	10^{-3}	10^{-3}	3.03	50	102	51

The **scd** values represents the minimum number of significant correct digits in the numerical solution at the end of the integration interval, i.e.

$$\mathbf{scd} = -\log(\| \text{ relative error at the end of the integration interval } \|_\infty)$$

Numbers of right part evaluations and decompositions are denoted by **f** and **dec**, respectively. At that, right parts computed to get numerical Jacobi matrices are not taken into account.

Conclusion

The greatest efficiency of the derived algorithm is reached on performing calculations with low accuracy $\varepsilon \le 10^{-2}$. This is due to low order of numerical formulas involved in solver iros2. The efficiency of new algorithm may be increased through freezing the Jacobi matrix (i.e. applying same Jacobi matrix over several integration steps). This can reduce computational costs due to evaluating and decomposing the Jacobi matrix. Furthermore, note that deriving reliable non-iterative algorithms that have high accuracy and efficiency is possible using methods based on (m, k)-methods of high order [9].

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