Numerical Simulations of Reaction-Diffusion Systems Arising in Chemistry Using Exponential Integrators

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Abstract. We perform a comparative numerical study of two reaction-diffusion models arising in chemistry by using exponential integrators. The aim of exponential integrators is to handle exactly the linear part of the differential model and allow the remaining part of the integration to be integrated numerically using an explicit scheme. Numerical simulations of the reaction kinetics associated with these models, including both the local and global errors as a function of time step and error as a function of computational time are shown.

1 Introduction

The reaction-diffusion models can display a wide variety of spatio-temporal phenomena in response to large amplitude perturbations. The nature of the kinetic terms plays the important role in determining the solution behaviour. The efficient and accurate simulation of such systems, however, represent a difficult task. This is because they couple a stiff diffusion term with a (typically) strongly nonlinear reaction term. When discretised this leads to large systems of strongly nonlinear, stiff ODEs.

In this work we carry out a comparative numerical approach of two reaction-diffusion models arising in chemistry by using exponential integrators. The paper is organized as follows. Section 2 briefly describes the exponential integrators and their features. In Section 3 the two reaction kinetics – the chlorite-iodide-malonic acid (CIMA) starch reaction and a glycolysis model governed by a cross kinetics, respectively – are presented on the basis of which the numerical study is carried out. Section 4 is devoted to a short description of the numerical schemes applied to the models under study, together with results of the numerical simulations. Some concluding remarks are drawn in the last section.

2 General Framework of the Exponential Integrators

The exponential integrators represent numerical schemes specifically constructed for solving differential equations (see for details [11]), where it is possible to split

the problem into a linear and a nonlinear part

$$\dot{y} = \mathcal{L}y + \mathcal{N}(y,t), \qquad y(t_{n-1}) = y_{n-1}, \tag{1}$$

where $y \in \mathbf{C}^d$, $\mathcal{L} \in \mathbf{C}^{d \times d}$ and $\mathcal{N} : \mathbf{C}^d \times \mathbf{R} \to \mathbf{C}^d$. In specific applications (discretizations of PDEs) the matrix $\mathcal{L}$ is unbounded. Generally, solving such problems requires an implicit scheme; the goal of the exponential integrators is to treat the linear term exactly and allow the remaining part of the integration to be integrated numerically using an explicit scheme.

An exponential integrator has two main characteristics: (i) If $\mathcal{L} = 0$, then the scheme reduces to a standard general linear scheme. This is often called the underlying general linear scheme; (ii) If $\mathcal{N}(y,t) = 0$ for all y and t, then the scheme reproduces the exact solution of (1). To satisfy (i) the exponential function must be used within the numerical scheme. Despite the fact that $\mathcal{L}$ is unbounded, typically the coefficients of the scheme will be bounded.

For an s-stage exponential integrator of Runge-Kutta type, we define the internal stages and output approximation:

$$Y_i = h \sum_{j=1}^{s} a_{ij}(h\mathcal{L})\mathcal{N}(Y_j, t_{n-1} + c_j h) + u_{i1}(h\mathcal{L})y_{n-1}, \quad i = 1, \ldots, s,$$

$$y_n = h \sum_{i=1}^{s} b_i(h\mathcal{L})\mathcal{N}(Y_j, t_{n-1} + c_j h) + v_1(h\mathcal{L})y_{n-1}. \tag{2}$$

The feature (i) above is satisfied if we require in (2) as $u_{i1}(0) = 1$, $a_{ij}(0) = a_{ij}$, $v_1(0) = 1$, and $b_i(0) = b_i$, where the real numbers a_{ij} and b_j represent the coefficients of the underlying Runge-Kutta scheme.

A step of length h in an exponential general linear scheme, requires to import r aproximations into the step, denoted as $y_i^{[n-1]}$, $i = 1, \ldots, r$. The internal stages (as in the Runge-Kutta case) are written as Y_i, $i = 1, \ldots, s$. After the step is completed, r updated approximations are computed. These are then used in the next step. Each step in an exponential general linear scheme can be written as

$$Y_i = h \sum_{j=1}^{s} a_{ij}(h\mathcal{L})\mathcal{N}(Y_j, t_{n-1} + c_j h) + \sum_{j=1}^{r} u_{ij}(h\mathcal{L})y_j^{[n-1]}, \quad i = 1, \ldots, s,$$

$$y_i^{[n]} = h \sum_{j=1}^{s} b_{ij}(h\mathcal{L})\mathcal{N}(Y_j, t_{n-1} + c_j h) + \sum_{j=1}^{r} v_{ij}(h\mathcal{L})y_j^{[n-1]}, \quad i = 1, \ldots, r. \tag{3}$$

The exponential integrators of Runge-Kutta type are easily seen to be a special case when $r = 1$ with $u_{i1}(z) = a_{i0}(z)$, $v_{11}(z) = b_0(z)$ and $b_{1j}(z) = b_j(z)$.

3 Defining the Model Equations

In this section we shortly describe the models governing different reaction kinetics arising in chemistry, which will be solved numerically in Section 4. Following

nondimensionalization of the system in space, such that the spatial domain becomes $[0, 1]$, the general system is represented by the following equations:

$$u_t = D_u u_{xx} + f(u, v), \qquad v_t = D_v v_{xx} + g(u, v),$$

where u and v are the concentrations of the two morphogens, D_u and D_v are the corresponding diffusion coefficients, and $f(u, v)$ and $g(u, v)$ encode the reaction kinetics between u and v.

Model describing the chlorite-iodide-malonic acid starch reaction. The first Turing patterns were observed in the chlorite-iodide-malonic acid starch reaction (CIMA) reaction ([7]). The model proposed by Lengyel and Epstein ([10]) stresses three processes: the reaction between malonic acid (MA) and iodine to create iodide, and the reactions between chlorite and iodide and chloride and iodide. The form of these reactions is indicated in [6].

By imposing the experimentally realistic hypothesis that the concentration of malonic acid, chlorine dioxide and iodine are constant, Lengyel and Epstein obtained the following model:

$$\frac{\partial u}{\partial t} = k_1 - u - \frac{4uv}{1 + u^2} + \nabla^2 u, \qquad \frac{\partial v}{\partial t} = k_2 \left[k_3 \left(u - \frac{uv}{1 + u^2} \right) + c\nabla^2 v \right],$$

where u, v are the concentrations of iodide and chlorite, respectively and k_1, k_2, k_3 and c are positive constants.

Glycolysis model with cross kinetics. We also analyze numerically a glycolysis model ([1]) with cross kinetics (where the activator upregulates itself, but downregulates the inhibitor, and the inhibitor likewise upregulates the activator and downregulates itself). In this model, the functions f and g are given by

$$f(u, v) = ru^2v + \nu v - \mu u, \qquad g(u, v) = r(1 - u^2v) - \nu v.$$

Here, the parameters r, μ, and ν are positive constants.

4   Numerical Schemes and Computational Issues

Here, we present the numerical schemes defining the exponential integrators that have been used in our simulations. All these integrators belong to the package `EXPINT` written in Matlab ([2]). In this description we will use two terms of order. The non-stiff order refers to the case when the operator $\mathcal{L}$ is bounded, such conditions were derived in [11]. The stiff order refers to the case when $\mathcal{L}$ is unbounded ([4]), for various schemes.

The first scheme that has been applied to our models is named `Lawson4`. The scheme `Lawson4` belongs to the Lawson schemes constructed by applying the Lawson transformations ([9]) to the semi-linear problem. It is based on the clasical fourth order scheme of Kutta (see [3], Eq. (235i)), and this scheme has stiff order one.

The scheme denoted by `hochost4` was developed by Hochbruck and Ostermann. It has five-stages and is the only known exponential Runge-Kutta method with stiff order four.
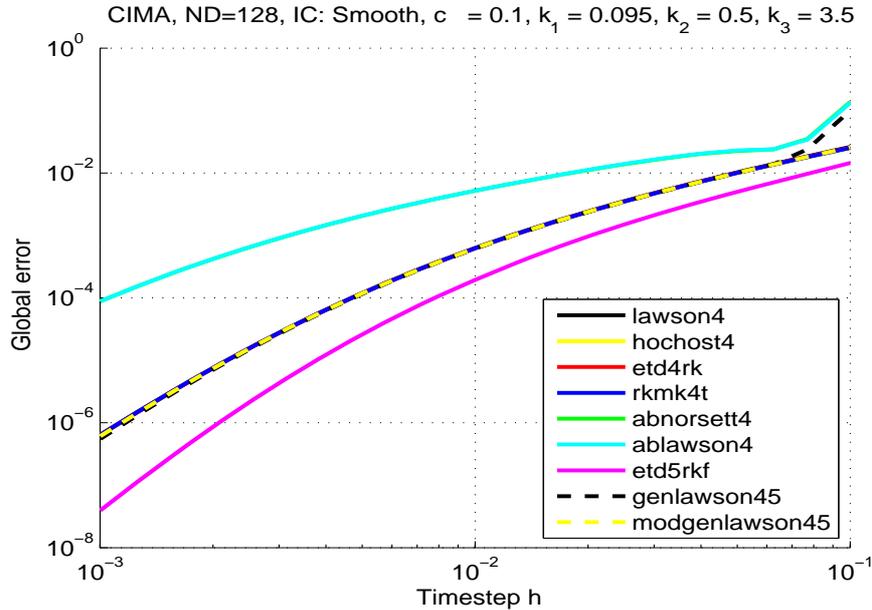
Fig. 1. Comparative results concerning the quality of the numerical schemes: the global error as a function of timestep h for the CIMA reaction kinetics.

Nørsett designed in [13] a class of schemes which reduced to the Adams-Bashforth methods when the linear part of the problem is zero.

ABLawson4 has stiff order one and is based on the Adams-Bashforth scheme of order four and is represented in this way so that the incoming approximation has the form $y^{[n-1]} = [y_{n-1}, hN_{n-2}, hN_{n-3}, hN_{n-4}]^T$.

ABNørsett4 is a stiff order four scheme of Norsett ([13]), which is implemented so that the incoming approximation has the same form as in ABLawson4.

ETD schemes are based on algebraic approximations to the nonlinear term in the variation of constants formula. ETD means "Exponential Time Differencing" and the name stems from [5]. The scheme ETD4RK due to Cox and Matthews in ([5], Eqs. (26)-(29)) started the recent focus on exponential integrators, unfortunately it has only stiff order two. ETD5RKF is a non-stiff fifth order scheme based on the six stage fifth order scheme of Fehlberg.

The scheme RKMK4t uses a convenient truncation of the $dexp^{-1}$ operator, leading to the method of Munthe-Kaas [12], which again is of stiff order two, but suffers from instabilities when non-periodic boundary conditions are used.

Krogstad [8] constructed the generalized Lawson schemes as a means of overcoming some of the undesirable properties of the Lawson schemes. This class of schemes uses approximations of the nonlinear term from previous steps, resulting in an exponential general linear method. The scheme genlawson45 included in the package mentioned above is also used for our numerical study.
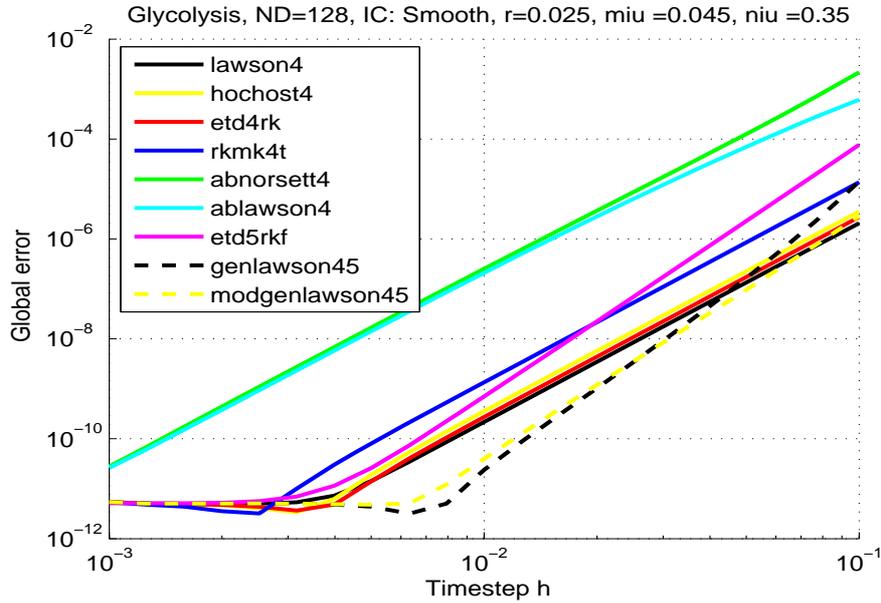
Fig. 2. Comparative results concerning the quality of the numerical schemes: the global error as a function of timestep h for the glycolysis model with cross reaction kinetics.

Figures 1 and 2 illustrate comparative results concerning the quality of the numerical schemes that have been used in this analysis. There are shown relationships between the global error and the timestep h varying from 10^{-3} to 10^{-1}. We note that for CIMA reaction model the best behaviour has had the scheme `etd5rkf`, while the scheme `ablawson4` indicated a significant increasing rate of the global errors with respect to the computed global error (see Fig. 1). Practically, we found the same behaviour for the other schemes implemented for this model. In the case of cross reaction kinetics for glycolysis model, the schemes `genlawson45` and `modgenlawson45` have had the best behaviour (see Fig. 2).

Figures 3 and 4 give timing results in the sense that present dependencies of the global error as a function of computational time for the two models. In this respect, good results are obtained with the schemes `lawson4`, `rkmk4t` and `etd5rkf` for the CIMA reaction model (see Fig. 3), and `lawson4`, `etd4rkf`, `rkmk4t` for the glycolysis model (see Fig. 4).

Figures 5 and 6 contain the results of a local order test applied for the two models under investigation. Here, other two exponential Runge-Kutta methods – Strehmel-Wiener and Friedli schemes, both with stiff order three – are added. This test does not make any sense for multistep-schemes. The vector of timesteps is built up, so that the smallest timestep must divide all other timesteps used. Otherwise, the computation would take a longer time.
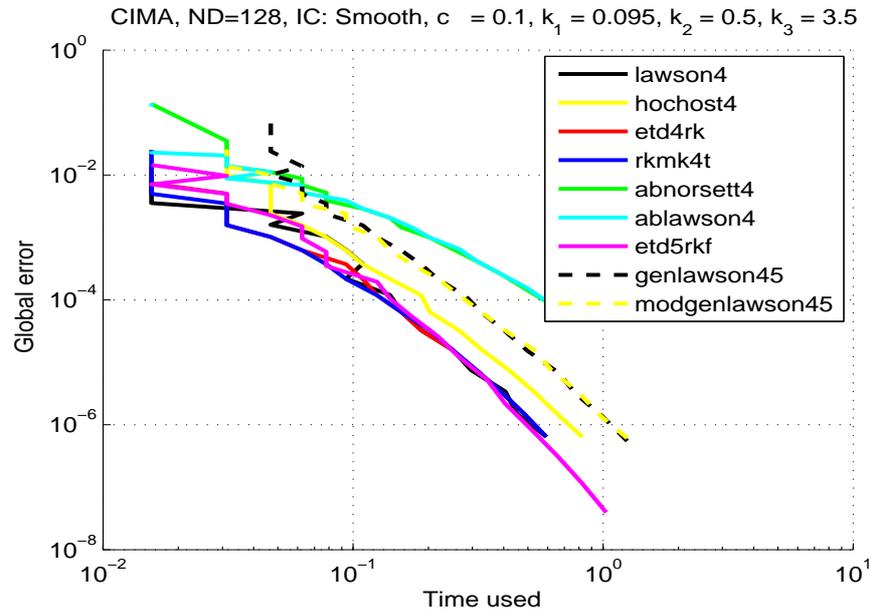
Fig. 3. Comparative results concerning the global error as a function of computational time for the CIMA reaction kinetics.

The plots precise in their title $ND = 128$ and "IC: Smooth". This means that we have used 128 Fourier modes in the spatial direction (must be power of 2), and as initial condition for the model variables, we have chosen a set of values with a Gaussian distribution.

5   Conclusions

In this study we carried out a comparative numerical approach for two models arising in chemistry: the chlorite-iodide-malonic acid (CIMA) starch reaction and a glycolysis model governed by a cross kinetics. The numerical results have been obtained by using several exponential integrators belonging to Matlab package EXPINT ([2]). Illustrative plots were presented concerning comparative global and local errors as well as timing results of the integration schemes.

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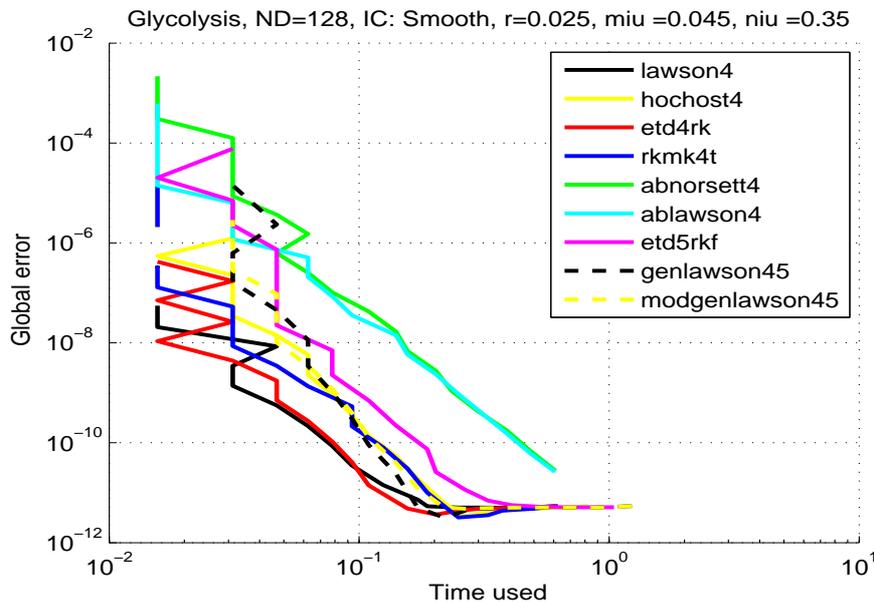3. Butcher, J.C.: *Numerical methods for ordinary differential equations.* John Wiley & Sons (2003).

Fig. 4. Comparative results concerning the global error as a function of computational time for the glycolysis model with cross reaction kinetics.

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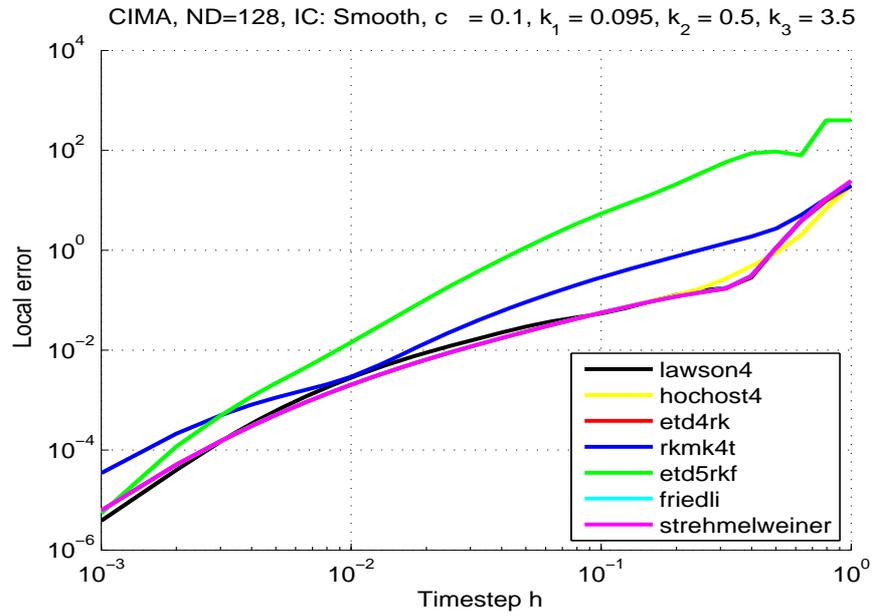
Fig. 5. Comparative results concerning the quality of the numerical schemes: the local error as a function of timestep h for the CIMA reaction kinetics.
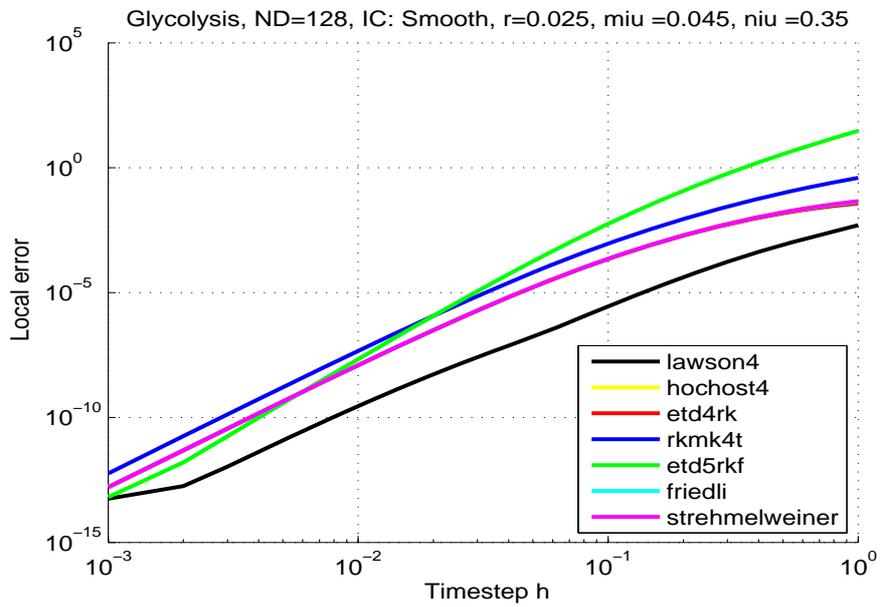

Fig. 6. Comparative results concerning the quality of the numerical schemes: the local error as a function of timestep h for the glycolysis model with cross reaction kinetics.