

A Lattice Boltzmann Method for the Chemical Clock in the Belousov–Zhabotinskii Reaction *

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A lattice Boltzmann method for the Belousov–Zhabotinskii reaction is proposed to simulate the chemical clock. Applying the Chapman–Enskog expansion and multiscale technique, we obtain a series of lattice Boltzmann equations in different timescales, the conservation law in timescale t_0 and coefficients of macroscopic equations to find the equilibrium distribution functions. A simple numerical scheme is designed to simulate the diffusion–reaction systems. The numerical example shows that the proposed method can be used to simulate chemical systems with the chemical clock.

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In the past decade, the lattice Boltzmann method (LBM) has attracted great attention as an alternative numerical method for simulation of fluid flows.^[3–5] The main idea of the LBM is to obtain available macroscopic physical equations by using the discrete Bhatnagar–Gross–Krook-type Boltzmann equations.^[3] Generally, one uses the equilibrium distribution function to fit some requirements which can be obtained with the multiscale technique and Chapman–Enskog expansion. Recently, there have been some studies that applied the LBM to reaction systems,^[6–8] and it is shown that the method is suitable for simulating chemical reactions. In this letter, we construct an appropriate LBM to simulate the Belousov–Zhabotinskii reactions, where there are six species (A , B , D , E , X and Y) and four step reactions:^[1,2]



The Belousov–Zhabotinskii reaction has a wide range of physical backgrounds and practical applications, not only in the area of nonlinear dynamics but also in other physical fields. For simulating this reaction system, we expand the distribution function to the order of three by the Chapman–Enskog expansion. Using the conservation law in the timescale t_0 , we obtain the error term of the equations reaching $O(\varepsilon^2)$, where ε is the Knudsen number. The numerical results show that our LBM is suitable for simulating the Belousov–Zhabotinskii reaction.

Let us discretize the velocity of particles into b directions in D -dimensional space. The lattice unit spacing is used where each node has b nearest neighbours connected by b links. The distribution function f_α^σ is the probability of finding a particle at time

t , node \mathbf{x} , with velocity \mathbf{e}_α , where $\alpha = 0, 1, \dots, b$ ($\alpha = 0$ is a rest particle). For a one-dimensional lattice, $D = 1$, $b = 2$, the particle velocities are $\mathbf{e}_\alpha = (0, c, -c)$; for a two-dimensional (2D) lattice, $D = 2$ and $b = 4$ leads to 2D square models. The macroscopic quantity u is defined by

$$u^\sigma = \sum_{\alpha} f_{\alpha}^{\sigma}(\mathbf{x}, t), \quad (1)$$

and the conservation condition is

$$\sum_{\alpha} f_{\alpha}^{\sigma, \text{eq}}(\mathbf{x}, t) = u^{\sigma}(\mathbf{x}, t), \quad (2)$$

where $\sigma = 1, 2, \dots, M$, u^σ is the density of the species σ at time t and position \mathbf{x} , and M is the number of species. Here we take $M = 2$.

The lattice Boltzmann equation reads^[5,6]

$$f_{\alpha}^{\sigma}(x + \Delta x, t + \Delta t) - f_{\alpha}^{\sigma}(x, t) = \Omega_{\alpha}^{\sigma}(x, t) + \omega_{\alpha}^{\sigma}(x, t). \quad (3)$$

In the case of the standard lattice Boltzmann,^[3–5] the term $\omega_{\alpha}^{\sigma} = 0$, where $\Omega_{\alpha}^{\sigma} = -(1/\tau)[f_{\alpha}^{\sigma}(\mathbf{x}, t) - f_{\alpha}^{\sigma, \text{eq}}(\mathbf{x}, t)]$, $f_{\alpha}^{\sigma, \text{eq}}$ is the equilibrium distribution function at t and \mathbf{x} ; ω_{α}^{σ} is the additional term; τ is the single relaxation time factor.

The small parameter ε is selected as the time unit in the numerical simulation, and it can also play the role of the Knudsen number.^[9] The lattice Boltzmann equation in physical units is

$$f_{\alpha}^{\sigma}(x + \varepsilon e_{\alpha}, t + \varepsilon) - f_{\alpha}^{\sigma}(x, t) = -\frac{1}{\tau}[f_{\alpha}^{\sigma} - f_{\alpha}^{\sigma, \text{eq}}] + \omega_{\alpha}^{\sigma}. \quad (4)$$

We apply the Taylor expansion, Chapman–Enskog expansion,^[10] and multiscale technique to Eq. (4), and assume

$$\omega_{\alpha}^{\sigma} = \varepsilon^2 \phi_{\alpha}^{\sigma}, \quad (5)$$

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then a series of lattice Boltzmann equations in different timescales are obtained:^[11]

$$\frac{\partial f_{\alpha}^{\sigma,(0)}}{\partial t_0} + e_{\alpha} \frac{\partial f_{\alpha}^{\sigma,(0)}}{\partial x} = -\frac{1}{\tau} f_{\alpha}^{\sigma,(1)}, \quad (6)$$

$$\frac{\partial f_{\alpha}^{\sigma,(0)}}{\partial t_1} - \tau \left(1 - \frac{1}{2\tau}\right) \left(\frac{\partial}{\partial t_0} + e_{\alpha} \frac{\partial}{\partial x}\right)^2 f_{\alpha}^{\sigma,(0)} = -\frac{1}{\tau} f_{\alpha}^{\sigma,(2)} + \phi_{\alpha}^{\sigma}, \quad (7)$$

$$\begin{aligned} & \frac{\partial f_{\alpha}^{\sigma,(0)}}{\partial t_2} + (1 - 2\tau) \left(\frac{\partial}{\partial t_0} + e_{\alpha j} \frac{\partial}{\partial x_j}\right) \frac{\partial f_{\alpha}^{\sigma,(0)}}{\partial t_1} \\ & + \left(\tau^2 - \tau + \frac{1}{6}\right) \left(\frac{\partial}{\partial t_0} + e_{\alpha j} \frac{\partial}{\partial x_j}\right)^3 f_{\alpha}^{\sigma,(0)} \\ & = -\frac{1}{\tau} f_{\alpha}^{\sigma,(3)} + (-\tau) \left(\frac{\partial}{\partial t_0} + e_{\alpha j} \frac{\partial}{\partial x_j}\right) \phi^{\sigma}(u^{\sigma}). \end{aligned} \quad (8)$$

Taking the summation over α in Eqs. (6) and (7), and performing (6)+(7) $\times \varepsilon$, we have

$$\frac{\partial u^{\sigma}}{\partial t} = \lambda^{\sigma} \varepsilon \left(\tau - \frac{1}{2}\right) \frac{\partial^2 u^{\sigma}}{\partial x_i \partial x_i} + \varepsilon \phi^{\sigma}(u^{\sigma})(b+1) + O(\varepsilon^2). \quad (9)$$

If we choose

$$\sum_{\alpha} f_{\alpha}^{\sigma,(0)} e_{\alpha i} = 0, \quad (10)$$

$$\sum_{\alpha} f_{\alpha}^{\sigma,(0)} e_{\alpha i} e_{\alpha j} = \lambda^{\sigma} u^{\sigma} \delta_{ij}, \quad (11)$$

where $\lambda^{\sigma} = D_{\sigma}/\varepsilon(\tau - 1/2)$, then Eq. (9) has the truncation error

$$R = O(\varepsilon^2). \quad (12)$$

By simple algebraic operation, we obtain equilibrium distribution functions $f_{\alpha}^{\sigma,(0)}$ as follows:

$$f_0^{\sigma,(0)} = u^{\sigma} - \frac{\lambda^{\sigma} D u^{\sigma}}{c^2}, \quad (13)$$

$$f_{\alpha}^{\sigma,(0)} = \frac{\lambda^{\sigma} D u^{\sigma}}{b c^2}, \alpha = 1, \dots, b. \quad (14)$$

From Eq. (9), we obtain the following diffusion-reaction equation (M=2, $u^1 = u$, $u^2 = v$),

$$\begin{aligned} \frac{\partial u}{\partial t} &= D_1 \nabla^2 u + [A - (B+1)u + u^2 v], \\ \frac{\partial v}{\partial t} &= D_2 \nabla^2 v + [Bv - u^2 v], \end{aligned} \quad (15)$$

which can be derived from the Belousov-Zhabotinskii reaction.

Next, we select the parameters as $A = 2.0$, $B = 5.45$ and $L = 1.0$ for the reaction terms. The diffusion coefficients are $D_1 = 8.0 \times 10^{-3}$ and $D_2 = 4.0 \times 10^{-3}$. The parameters in the lattice Boltzmann equation are: $c = 3.0$, $1/\tau = 1.2$, $b = 2$. These additional terms can be given as follows

$$\phi^1 = \frac{1}{\varepsilon(b+1)} [A - (B+1)u + u^2 v],$$

$$\phi^2 = \frac{1}{\varepsilon(b+1)} [Bv - u^2 v],$$

$$\lambda^1 = \frac{D_1}{\varepsilon(\tau - 1/2)}, \quad \lambda^2 = \frac{D_2}{\varepsilon(\tau - 1/2)}.$$

The boundary conditions can be expressed as $u|_{x=0,L} = A$ and $v|_{x=0,L} = B/A$ together with the initial values of

$$u(x, 0) = 2 - \sin(\pi x), \quad v(x, 0) = \frac{B}{A} + \varepsilon.$$

We also simulate this test problem using a finite difference method (FDM) to compare with the LBM results. In the FDM, Eq. (15) is solved with a time step $\Delta t_{\text{FDM}} = 1/12$, and a spatial step $\Delta x_{\text{FDM}} = L/N$. We find that the LBM results agree very well with the FDM results. In Fig. 1, the evolution of time is from $t = 5000\Delta t$ to $t = 15000\Delta t$. The numerical results show a periodic oscillation when $t > 6000\Delta t$, with a period of $2000\Delta t$. Figure 2 shows the wave and propagation of species u from time $t = 6000\Delta t$ to $t = 8000\Delta t$. It is noted that the chemical clock as computed using the LBM is in close agreement with that obtained using the FDM.

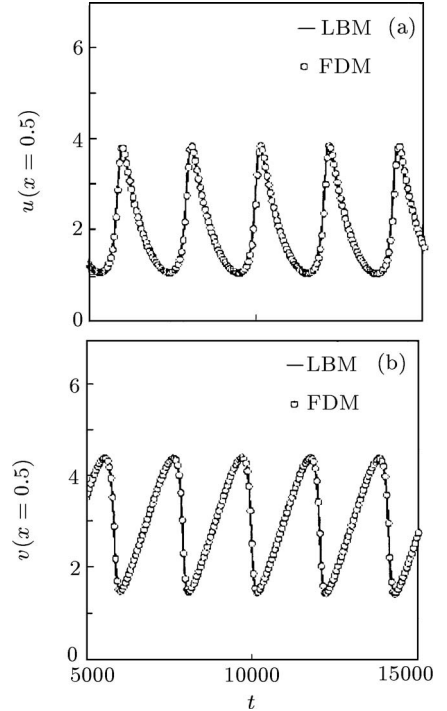


Fig. 1. LBM (line) and FDM (circles) results of u and v at $x = 0.5$ versus time t in the Belousov-Zhabotinskii reaction for the parameters, $A = 2.0$, $B = 5.45$ and $L = 1.0$, and the diffusion coefficients, $D_1 = 8.0 \times 10^{-3}$ and $D_2 = 4.0 \times 10^{-3}$. The parameters appearing in the lattice Boltzmann equation are: $c = 3.0$, $\tau = 1/1.2$, $b = 2$, $\Delta x = L/N$, $N=200$, $\varepsilon = \Delta t = \Delta x/c = 1/600$. In the FDM, the time step is $\Delta t_{\text{FDM}} = 1/12$, and the spatial step is $\Delta x_{\text{FDM}} = L/N$.

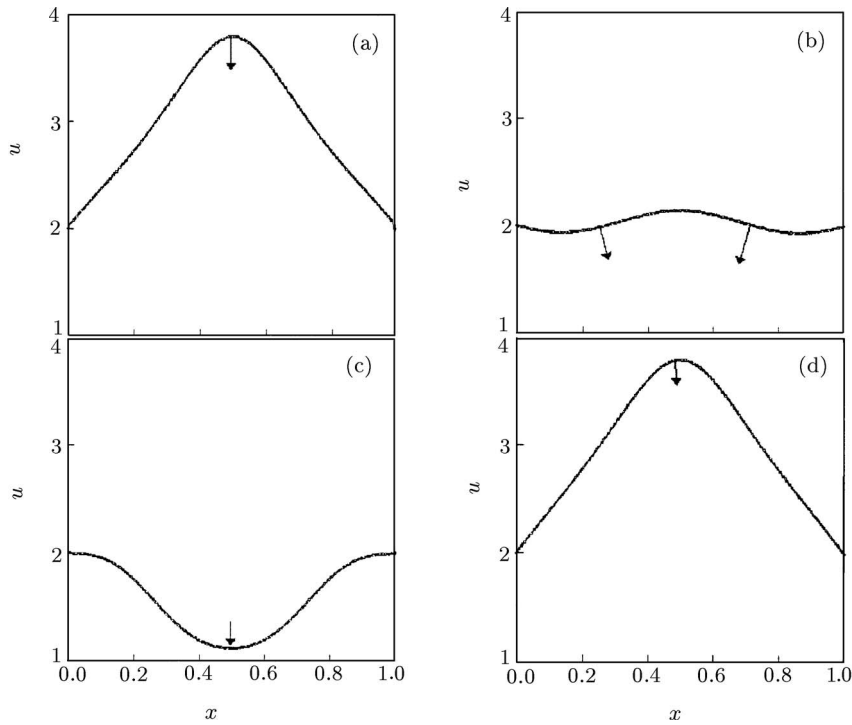


Fig. 2. Chemical clock in the Belousov–Zhabotinskii reaction. These curves show wave and propagation of species $u(x, t)$ at four time steps: (a) $t = 6000\Delta t$, (b) $t = 6500\Delta t$, (c) $t = 7500\Delta t$ and (d) $t = 8000\Delta t$. The parameters used are $A = 2.0$, $B = 5.45$ and $L = 1.0$. The diffusion coefficients are $D_1 = 8.0 \times 10^{-3}$ and $D_2 = 4.0 \times 10^{-3}$. The parameters appearing in the lattice Boltzmann equation are: $c = 3.0$, $\tau = 1/1.2$, $b = 2$, $\Delta x = L/N$, $N = 200$, $\varepsilon = \Delta t = \Delta x/c$. The arrow in the plot represents the movement direction of the wave.

In conclusion, we have developed a lattice Boltzmann method for simulating the Belousov–Zhabotinskii reaction, and show that the method can capture the well-known chemical clock of the diffusion-reaction system. Although we only studied the Belousov–Zhabotinskii reaction as an example, it is easy to construct other nonlinear chemical reaction systems by using higher moments of the lattice Boltzmann method. This method may be a viable tool to simulate some patterns of chemical reactions. A key assumption we used in this model is $\omega_\alpha^\sigma = \varepsilon^2 \phi_\alpha^\sigma$, which may be useful in other applications. The meaning of this assumption is that reaction and diffusion have the same order of scales. Our numerical results show that the assumption is reasonable. Until now, there has been no method that does not use this assumption in the field of the reaction-lattice Boltzmann method. As a numerical method, the lattice Boltzmann method may not be more efficient and more accurate than standard FDM schemes. However, we do find the ability of the LBM to simulate the nonlinear systems and the diffusion-reaction systems.

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