

Nonlinear reactions advected by a flow

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Abstract

Flows can modify the effects of nonlinear reactions. Here we use a lattice Boltzmann method to simulate an athermal flow which advects reactant species. As an example we consider a reactive system modeled by the Brusselator which exhibits Turing structures; the system is subjected to mixing as induced by the wakes behind obstacles. The resulting effect is a modification of the reaction patterns which can be as dramatic as their disappearance.

1 Introduction

The lattice Boltzmann method is a numerical method developed for simulating hydrodynamics [1] that was derived from lattice gas automaton methods [3]. In these methods, particles propagate on a lattice with discrete velocities and collide on the lattice sites. The lattice Boltzmann method replaces the particles with particle densities, which are propagated according to the discrete velocities. The collision step is replaced by a relaxation of the particle densities towards their equilibrium value as defined by an equilibrium distribution which depends only on the hydrodynamic quantities (mass and momentum). By a proper choice of this equilibrium distribution it is possible to avoid the problems of the dependence on the lattice geometry and of non-galilean invariance, which plague the lattice gas methods.

The lattice Boltzmann method for fluid dynamics is used here to simulate a solvent which carries reactants. We assume that reactant concentrations are sufficiently low to not influence the hydrodynamics. Thus the reactants take the role of passive scalars that are advected by the flow. Interaction between the reactive species is an additional process during collision. The fluid transport using the lattice Boltzmann method introduces a (numerical) diffusion

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of the advected species. This diffusion can be tuned by introducing additional diffusive steps for different reactants yielding different diffusion coefficients. The equations we simulate are for the solvent (n_0)

$$\frac{\partial n_0}{\partial t} + (\nabla \cdot n_0 \mathbf{u}) = 0, \quad (1a)$$

$$\frac{\partial n_0 \mathbf{u}}{\partial t} + [\nabla \cdot n_0 \mathbf{u} \mathbf{u}] + \nabla p + [\nabla \cdot \boldsymbol{\tau}] = 0 \quad (1b)$$

with

$$\boldsymbol{\tau} = -\nu \left(\{\nabla \mathbf{u}\} + \{\nabla \mathbf{u}\}^T - \frac{2}{d} (\nabla \cdot \mathbf{u}) \boldsymbol{\delta} \right) - \mu (\nabla \cdot \mathbf{u}) \boldsymbol{\delta}, \quad (1c)$$

where p is the pressure, d the dimension of space, ν the kinematic viscosity and μ the bulk viscosity. The motion of the solvent advects the solutes or reactants (n_s) which obey the evolution equation

$$\frac{\partial n_s}{\partial t} + (\nabla \cdot n_s \mathbf{u}) = D_s \nabla^2 n_s + F_s(\{n_j\}), \quad (2)$$

where D_s is the diffusion coefficient for species s , and $F_s(\{n_j\})$ are nonlinear reactive terms that depend on the concentrations of all reactants.

2 Lattice Boltzmann method

We use a lattice Boltzmann method [4–6,9,10] to obtain the equations of fluid dynamics for the solvent. The universe consists of a regular lattice, on which particles propagate with discrete velocities \mathbf{e}_i . The velocity vectors are multiples of the lattice basis vectors, so that all particles stay on the lattice after propagating for one time step. The probability density of particles propagating from node \mathbf{x} at time t in direction i is $f_i(\mathbf{x}, t)$ (in short f_i). The procedure of the lattice Boltzmann method is based on the lattice Boltzmann equation

$$f_i(\mathbf{r} + \mathbf{e}_i, t + 1) - f_i(\mathbf{r}, t) = \Omega_i. \quad (3)$$

The collision term Ω_i expresses the change in occupation of channel i due to the collision with other particles on the node. In general it is a complicated function of the particle densities f_j on all channels. Here we are not concerned with modeling a lattice gas scheme, but only in recovering the Navier-Stokes equations. The simplest procedure replaces the whole spectrum of eigenvalues of the collision operator by one single eigenvalue, i.e., we use a single

relaxation-time method [2] where the collision term on the r.h.s. of Eq. (3) is set to

$$\Omega_i = -\omega (f_i - f_i^{\text{eq}}). \quad (4)$$

f_i^{eq} is an equilibrium distribution of the velocities which depends only on the local density

$$n = \sum_i f_i \quad (5a)$$

and momentum

$$n\mathbf{u} = \sum_i \mathbf{e}_i f_i. \quad (5b)$$

Through this dependence on *local density* and *local momentum*, the collision term Ω_i depends on all f_j 's. The factor ω is a relaxation parameter, which is the inverse of a characteristic relaxation time. For $\omega = 1$, Eq. (3) reduces to

$$f_i(\mathbf{r} + \mathbf{e}_i, t + 1) = f_i^{\text{eq}}(\mathbf{r}, t), \quad (6)$$

which means that in each time step the distribution relaxes to the local equilibrium. Elementary stability requirements impose that $\omega \in [0, 2]$. We will see that for ω close to 2, which corresponds to an over-relaxation process, the viscosity has a very low value, and therefore a high Reynolds number can be achieved.

From the equations (3) and (4) describing the lattice Boltzmann method, one can derive the Navier-Stokes equations using the usual Chapman-Enskog expansion, and one obtains [10] the following conditions for the equilibrium distribution $f_i^{\text{eq}}(\mathbf{r}, t)$

$$\sum_i f_i^{\text{eq}} = n \quad (7a)$$

$$\sum_i \mathbf{e}_i f_i^{\text{eq}} = n\mathbf{u} \quad (7b)$$

$$\sum_i \mathbf{e}_i \mathbf{e}_i f_i^{\text{eq}} = n\mathbf{u}\mathbf{u} + p\boldsymbol{\delta}; \quad (7c)$$

$$\sum_i (\mathbf{e}_i \mathbf{e}_i \mathbf{e}_i)_{\alpha, \beta, \gamma} f_i^{\text{eq}} = n\mathbf{u}_\alpha \mathbf{u}_\beta \mathbf{u}_\gamma + p(\mathbf{u}_\alpha \delta_{\beta\gamma} + \mathbf{u}_\beta \delta_{\alpha\gamma} \mathbf{u}_\gamma \delta_{\alpha\beta}). \quad (7d)$$

One also obtains the shear viscosity $\nu = p \left(\frac{1}{\omega} - \frac{1}{2} \right)$ and bulk viscosity $\mu = \left(\frac{2}{d}p + p - \frac{\partial p}{\partial n} \right) \left(\frac{1}{\omega} - \frac{1}{2} \right)$, which reduces to $\mu = \frac{2}{d}p \left(\frac{1}{\omega} - \frac{1}{2} \right)$ when the equation

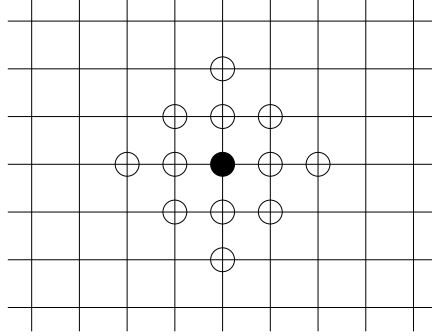


Fig. 1. Geometry of the velocity set for the solvent

of state is that of an ideal gas $p = c_s^2 n$.

To complete the description of the lattice Boltzmann method, we need to specify the lattice by the vectors \mathbf{e}_i and the equilibrium distribution. The model used in our simulations [10] employs 13 velocities on a square lattice (see Figure 1)

$$\mathbf{e}_0 = (0, 0) \tag{8a}$$

$$\mathbf{e}_{1..4} = (\pm 1, 0), (0, \pm 1) \tag{8b}$$

$$\mathbf{e}_{5..8} = (\pm 1, \pm 1) \tag{8c}$$

$$\mathbf{e}_{9..12} = (\pm 2, 0), (0, \pm 2). \tag{8d}$$

The local equilibrium is described by a polynomial expansion in the velocity \mathbf{u}

$$f_i^{\text{eq}} = n(A_i + B_i(\mathbf{e}_i \cdot \mathbf{u}) + C_i(\mathbf{e}_i \cdot \mathbf{u})^2 + D_i|\mathbf{u}|^2 + E_i(\mathbf{e}_i \cdot \mathbf{u})^3 + F_i(\mathbf{e}_i \cdot \mathbf{u})|\mathbf{u}|^2) \tag{9}$$

where n is the average density per node. The conditions imposed on the equilibrium distribution, together with the condition that the ratio between A_i and B_i be the same for all i , lead to the following relations:

$$\begin{aligned} A_i &= T_i & T_0 &= \frac{36}{96} \\ B_i &= 2T_i & T_{1..4} &= \frac{8}{96} \\ C_i &= 2T_i & T_{5..8} &= \frac{6}{96} \\ D_i &= -T_i & T_{9..12} &= \frac{1}{96} \\ E_{1..4} &= \frac{32}{96} & E_{5..8} &= \frac{12}{96} & E_{9..12} &= \frac{1}{96} \\ F_{1..4} &= -\frac{48}{96} & F_{5..8} &= -\frac{12}{96} & F_{9..12} &= 0. \end{aligned} \tag{10}$$

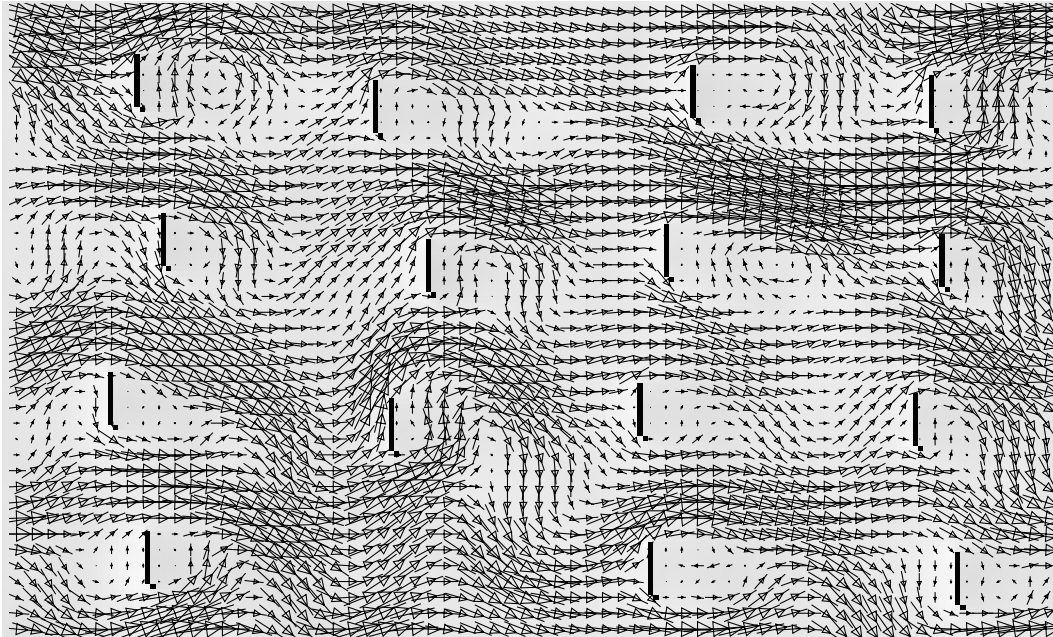


Fig. 2. Velocity field resulting from the flow around obstacles. The viscosity is $\nu = 0.01316$ and the average velocity is $\mathbf{u}_x = 0.05$.

This model has the equation of state $p = n/2$, which yields a sound speed of $c_s = \sqrt{1/2}$.

Using this lattice Boltzmann method, we set up the following flow to simulate strong mixing of a fluid: The fluid is forced uniformly such that the global average velocity is $u_x = 0.05$ (lattice units per time step). The fluid is forced past an array of fixed obstacles, which simply reflect back incoming fluid particles (and thus enforce zero fluid velocity). The effect is that a nonlaminar flow with many vortices develops as interacting wakes behind the obstacles. An instantaneous snapshot of the flow pattern is shown in Figure 2.

3 Advection mechanism

For the advection of the reactant species, there are two possible algorithms. One is to use the same velocity set for the reactants as for the solvent, and to use a similar relaxation mechanism together with an equilibrium distribution related to that of the solvent [7]. Here we use another method [10], where the velocity set for the reactants is much smaller (only five velocities $\mathbf{c}_0 = (0, 0)$ and $\mathbf{c}_{1..4} = (\pm 1, 0), (0, \pm 1)$). We distribute the available mass of reactant s ,

⬆ ⬇ ⬆	
→	$D + \frac{u_x}{2}$
↑	$D + \frac{u_y}{2}$
←	$D - \frac{u_x}{2}$
↓	$D - \frac{u_y}{2}$
•	$1 - 4D$

Fig. 3. Velocity configuration for the reactant species and weights depending on the diffusion coefficient D and the velocity $\mathbf{u} = (u_x, u_y)$.

$n_s = \sum_i f_i^s$, onto these channels according to

$$f_i^s(\mathbf{r} + \mathbf{c}_i, t + 1) = \begin{cases} (1 - 4D_s)n_s & \text{for } i = 0; \\ (D_s + \frac{1}{2}\mathbf{c}_i \cdot \mathbf{u})n_s & \text{for } i \neq 0. \end{cases} \quad (11)$$

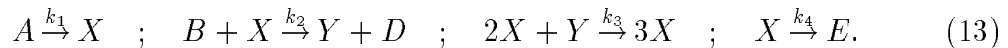
To avoid instabilities, all coefficients should be positive and thus $\frac{1}{2}|\mathbf{u}| < D_s < \frac{1}{4}$. A further increased diffusion coefficient for some species (as required to obtain Turing patterns) can be achieved by inserting one or more steps of pure diffusion without advection for this species:

$$f_i^s(\mathbf{r} + \mathbf{c}_i, t + 1) = \begin{cases} (1 - 4\tilde{D}_s)n_s & \text{for } i = 0; \\ \tilde{D}_s n_s & \text{for } i \neq 0. \end{cases} \quad (12)$$

The velocity sets and the distribution weights for the reactive species are shown in Figure 3.

4 Autocatalytic multimolecular reactive system

As an example of a nonlinear reactive system we use the Brusselator model [8,11]



We assume that species A, B, D, E are in abundance and held at constant concentration and only species X and Y are of interest. Then this model can be described after rescaling by the following reaction-diffusion equations with advection term

$$\partial_t x + (\nabla \cdot x \mathbf{u}) = D_x \nabla^2 x + A - (B + 1)x + x^2 y; \quad (14a)$$

$$\partial_t y + (\nabla \cdot y \mathbf{u}) = D_y \nabla^2 y + Bx - x^2 y. \quad (14b)$$

This model exhibits Turing patterns for the parameters $A = 2.5$, $B = 10$, $\delta = D_y/D_x = 3$.

The reactive dynamics is introduced by modifying the densities of the reactants n_s at each point and each time step according to

$$n_x \leftarrow n_x + \Delta t (A - (B + 1)n_x + n_x^2 n_y); \quad (15a)$$

$$n_y \leftarrow n_y + \Delta t (Bn_x - n_x^2 n_y). \quad (15b)$$

Thus the complete dynamics for the reactant species is

Reaction

$$n_s(\mathbf{r}) \leftarrow n_s(\mathbf{r}) + \Delta t F_s(\{n_s(\mathbf{r})\}) \quad (16a)$$

Diffusion

$$f_i^s(\mathbf{r} + \mathbf{c}_i) \leftarrow \begin{cases} (1 - 4\tilde{D}_s) n_s(\mathbf{r}) & \text{for } i = 0 \\ \tilde{D}_s n_s(\mathbf{r}) & \text{for } i \neq 0 \end{cases} \quad (16b)$$

Summation

$$n_s(\mathbf{r}) \leftarrow \sum_i f_i^s(\mathbf{r}) \quad (16c)$$

Diffusion + Advection

$$f_i^s(\mathbf{r} + \mathbf{c}_i, t + 1) \leftarrow \begin{cases} (1 - 4D_s) n_s(\mathbf{r}) & \text{for } i = 0 \\ (D_s + \frac{1}{2} \mathbf{c}_i \cdot \mathbf{u}) n_s(\mathbf{r}) & \text{for } i \neq 0 \end{cases} \quad (16d)$$

Summation

$$n_s(\mathbf{r}) \leftarrow \sum_i f_i^s(\mathbf{r}), \quad (16e)$$

where in step (16a) the $F_s(\{n_s(\mathbf{r})\})$ refer to the reactive terms in Eq. (15) and steps (16b) and (16c) are only used for those species with an increased diffusion coefficient.

When a non-stationary flow advects the reactants, formation of any regular pattern is prevented by the mixing effect of the changing flow. In Figure 4 we compare snapshots of systems without flow and with a nonstationary flow created by inserting an array of obstacles into a homogeneous flow.

The mixing effect produced by the flow can be quantified by measuring the mean squared deviation from the average value of x . Fig. 5 shows that the flow

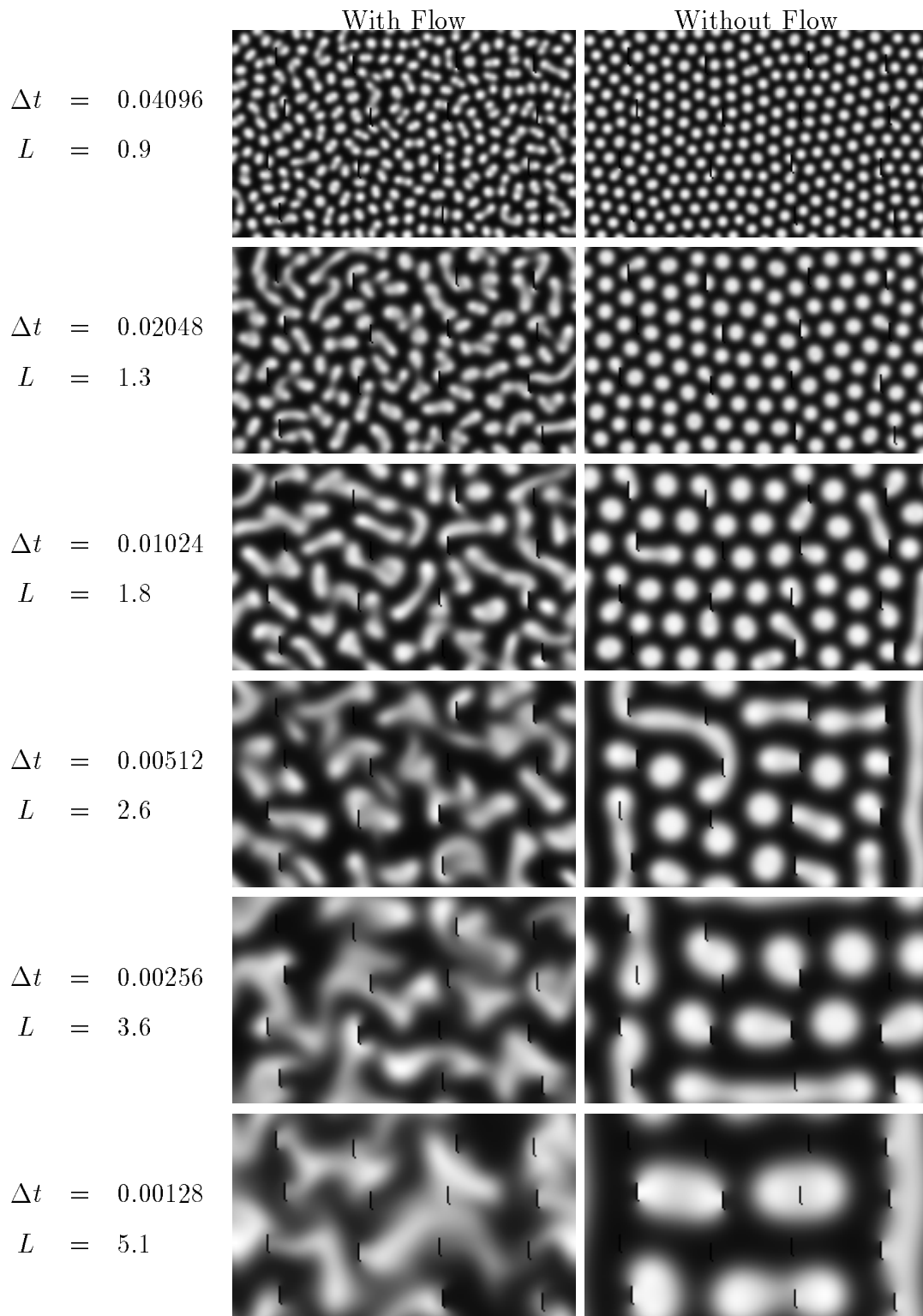


Fig. 4. Comparison of reaction with and without flow for different reactive time- and space-scales (L is the size of the Turing patterns relative to the size of the obstacles). The size is 200×120 nodes, with obstacles and flow as shown in Figure 2. The images show the variable x in a grey scale representation (white – high concentration).

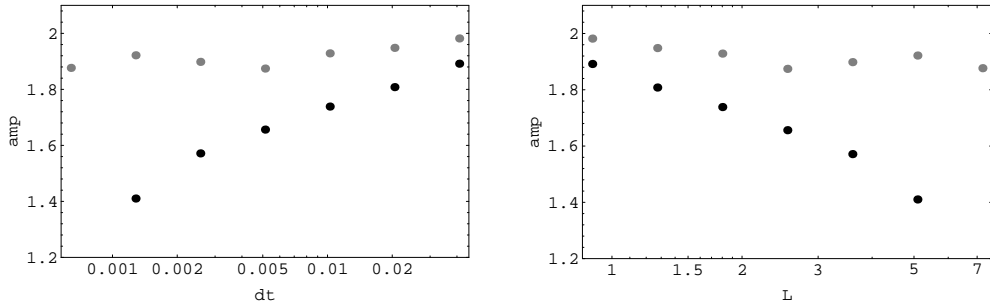


Fig. 5. Pattern amplitude (measured as standard deviation from the average concentration) for the reaction with flow and without flow as a function of Δt (on the left), and as a function of the relative size of the Turing patterns (on the right). The dark points show the situation with flow, and the grey points are measurements without flow.

reduces the pattern amplitude as the space scale of the patterns gets larger compared to the size of the obstacle. For large mixing (small obstacles) the system starts to oscillate homogeneously, just as it does when the diffusion coefficients for both x and y are equal.

5 Conclusion

We have developed a method for the simulation of the combined effects of an athermal flow (obeying the Navier-Stokes equations) with transport, reaction and diffusion of reactants. The simulation uses the lattice Boltzmann method with 13 velocities on a square lattice for simulating the flow and a smaller set of velocities for the reactive species. We have demonstrated the effect of turbulent mixing on pattern formation in the Brusselator model. Weak mixing prevents stationarity of the patterns, and strong mixing can destroy the patterns altogether. Further extensions of this work will include reciprocal effects of the reactant concentrations on the hydrodynamic flow.

References

- [1] R. Benzi, S. Succi, and M. Vergassola. The lattice Boltzmann equation: theory and applications. *Physics Reports*, 222(3):145–197, Dec. 1992.
- [2] P. L. Bhatnagar, E. P. Gross, and M. Krook. *Phys. Rev.*, 94:511, 1954.
- [3] Gary Doolen, editor. *Lattice gas methods for partial differential equations*, Redwood City, CA, 1990. Addison-Wesley.
- [4] F. Higuera, S. Succi, and R. Benzi. Lattice gas dynamics with enhanced collisions. *Europhys. Lett.*, 9(4):345–349, 1989.

- [5] J. M. V. A. Koelman. A simple lattice Boltzmann scheme for Navier-Stokes fluid flow. *Europhys. Lett.*, 15(6):603–607, 1991.
- [6] G. McNamara and Berni Alder. Analysis of the lattice Boltzmann treatment of hydrodynamics. *Physica A*, 194:218–228, 1993.
- [7] S. Ponce-Dawson, S. Chen, and G.D. Doolen. Lattice Boltzmann computations for reaction-diffusion equations. *Journal of Chemical Physics*, 98(2):1514–1523, 15 Jan. 1993.
- [8] I. Prigogine and R. Lefever. Symmetry breaking instabilities in dissipative systems. ii. *J. Chem. Phys.*, 48:1695–1700, 1968.
- [9] Y.H. Qian, D. d’Humières, and P. Lallemand. Lattice BGK models for Navier-Stokes equation. *Europhysics Letters*, 17(6):479–484, 1 Feb. 1992.
- [10] Jörg R. Weimar. *Cellular Automata Methods for Reactive Systems*. PhD thesis, Université Libre de Bruxelles, Belgium, 1995.
- [11] Anne De Wit. *Brisure de symétrie spatiale et dynamique spatio-temporelle dans les systèmes réaction-diffusion*. Thèse de doctorat, Université Libre de Bruxelles, Belgium, 1993.