Model for the Belousov-Zhabotinsky Reaction and Surface Flow Structure Induced by Chemical Concentration Gradients

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Abstract—The present paper proposes a model that explains propagating chemical waves and the surface flow velocity of the chemical solution in the Belousov-Zhabotinsky (BZ) reaction system. The model governing the chemical reaction is described by the reaction-diffusion equations having the non-linear reaction terms of the Oregonator model. The model combines with the displacement field of a spring network through the Marangoni effect and the convection terms. The model assumes that the surface of the chemical solution has the spatially connected spring network. The displacement of the spring network is caused by the Marangoni effect due to spatial concentration gradients of the chemical species; the displacement field also causes the transportation of the chemical species. Through numerical experiments, it is confirmed that the proposed model can partially explain the surface flow structure previously observed in the BZ reaction system.

1. Introduction

In the Belousov-Zhabotinsky (BZ) reaction system, a flow field is observed in a shallow layered chemical solution spread in a petri dish. The BZ reaction system organizes spatial patterns such as a target wave (a single wave) and a spiral wave (a wave train). Several researchers found that a single chemical wave or a wave train traveling in the shallow layered chemical solution induce surface flow fields and 3-dimensional flow structures [1, 2]. They have suggested that the Marangoni effect due to the spatial gradients of concentration distributions of the chemical species causes an inhomogeneous surface tension distribution and the surface tension distribution causes a flow field on the surface of the solution. However, the mechanism causing the flow field has not been clearly understood.

The essential mechanism of the BZ reaction system is described by a reaction-diffusion system. A particular point of the system performs as a non-linear oscillator expressing the non-linear chemical reaction. Diffusion on the chemical species connects spatially the non-linear oscillators. The reaction-diffusion system consisting of the oscil-

lators and the diffusion processes causes the spatial patterns of a target wave and a spiral one.

There are two major possible mechanisms causing a flow of chemical solution during chemical wave(s) propagation. One of the mechanisms is the Marangoni effect [2, 3]. The spatial concentration distribution of the chemical species causes a non-uniform distribution on the surface tension of the chemical solution. The Marangoni effect refers to the non-uniform surface tension distribution caused by the non-uniform concentration distribution of a chemical species. Therefore, the effect causes a flow field on the surface of chemical solution. In the BZ reaction system, a surface tension distribution due to a concentration distribution of chemical species have been observed. The other one is the gravity effect [4]. Chemical reaction waves cause a concentration distribution in the bulk of the chemical solution. Since the chemical species have different density, the gravity effect due to the density difference causes a flow of the chemical solution in the bulk. The flow in the bulk also causes a surface flow. However, according to these two types of the mechanisms, we can not explain perfectly the flow structure due to the chemical wave propagation. We need additional and/or novel mechanism for explaining the flow observed during the chemical wave propagation.

The present paper proposes a model for explaining the chemical wave propagation and the surface flow of the chemical solution. The model for the chemical reaction is the Oregonator model having the concentration of two types of chemical species: the activator and inhibitor variables and the convection terms along the surface of the solution. The model assumes that the surface of the solution has a spatially connected spring network and the displacement of the network is caused by the force due to the Marangoni effect, that is, the concentration gradient of a chemical species on the surface. Therefore, the model assumes that the chemical reaction system is coupled with the spring network through the Marangoni effect. Through 1-dimensional numerical experiments, we have confirmed that the model can explain partially the temporal changes of the surface flow velocity during a single wave propaga-

tion observed in the previous real laboratory experiments. Previous studies utilizing only the Marangoni effect and/or the gravity effect do not present such the flow structure observed on the surface of the BZ solution. In addition, we have also confirmed the surface flow structure of the chemical solution during wave train propagation.

2. Model

2.1. Chemical Reaction

The standard reaction-diffusion equations having the two variables $u(x,t)$ and $v(x,t)$ and their convection terms in the 1-dimensional domain of x are described by,

$$\frac{\partial u}{\partial t} = D_u \nabla^2 u + \frac{1}{\varepsilon} f(u,v) - V_x \frac{\partial u}{\partial x},$$
$$\frac{\partial v}{\partial t} = D_v \nabla^2 v + g(u,v) - V_x \frac{\partial v}{\partial x}, \qquad (1)$$

where D_u and D_v refer to the diffusion coefficients of u and v, and the functions $f(u,v)$ and $g(u,v)$ do to the reaction terms describing the non-linear oscillation. Equation (1) has the convection terms $V_x \partial u / \partial x$ and $V_x \partial v / \partial x$. The Neumann boundary condition on u and v is applied to both the edges of the 1-dimensional domain. The 1-dimensional domain is defined in $0 \le x \le L_x$.

The Oregonator model describes the reaction functions of $f(u,v)$ and $g(u,v)$ with the two variables u and v expressing concentration distributions of their corresponding chemical species,

$$f(u,v) = u(1-u) - pv \frac{u-q}{u+q},$$
$$g(u,v) = u - v. \qquad (2)$$

where p and q are constants. The BZ reaction system consists of Eqs. (1) and (2). The system under $D_u = D_v = 0$ and $V_x = 0$ becomes a single non-linear oscillator. The oscillator becomes an excitable system for $p < 0.5, 1+\sqrt{2} < p$ and an oscillatory system for $0.5 < p < 1+\sqrt{2}$.

The full reaction-diffusion system consisting of Eq.(1) and Eq.(2) without convection terms produces several types of spatio-temporal patterns. The excitable system generates a chemical wave by an external triggering process; the oscillatory system generates spontaneously a wave train at time intervals of its oscillating period. By triggering repeatedly the system or by breaking a chemical wave with an external force the excitable system also causes a wave train. These chemical waves travel in a shallow layer of the chemical solution spread in a petri dish.

2.2. Spatially Connected Spring Network

The BZ reaction system generates chemical waves and the concentration distribution of the chemical species on the surface of the chemical solution causes a surface tension distribution (the Marangoni effect). We here assume
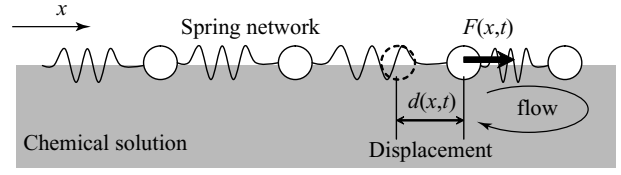

Figure 1: Spatially connected spring network. The variable $d(x,t)$ refers to the displacement of the solution surface from the equilibrium position. The function $F(x,t)$ refers to the force due to the Marangoni effect [see Eq.(5)].

that the non-uniform surface tension distribution induces a flow field on the chemical solution through a spatially connected spring network. The non-uniform surface tension induces a displacement of the network; the displacement causes a flow on the surface, as shown in Fig.1. The next equation describes the displacement field $d(x,t)$ of the spring network from the equilibrium position on a surface of the chemical solution,

$$\frac{\partial^2 d}{\partial t^2} + a \frac{\partial d}{\partial t} = D_d \nabla^2 d + F(x,t), \qquad (3)$$

where a and D_d are constants. The function $F(x,t)$ express the force due to the Marangoni effect at the position x. The temporal changes of the displacement becomes the velocity of the surface motion V_x.

$$V_x = \frac{\partial d}{\partial t} \qquad (4)$$

The Marangoni effect induces a surface tension distribution, which is caused by the spatial concentration gradient of a chemical species. When we assume that the distribution of the chemical species denoted by $v(x,t)$ causes the Marangoni effect, the force of Eq.(3) is defined by,

$$F(x,t) = b \frac{\partial v}{\partial x}. \qquad (5)$$

where b is a constant. The boundaries at the edge of the 1-dimensional system are fixed as $d(x = 0, t) = d(x = L_x, t) = 0$.

For numerical experiments, Eqs. (1)–(5) are discretized by the finite difference method with the spatial and temporal finite differences δx and δt. The Laplacian operator ∇^2 is discretized by the 5-points approximation method and the Crank-Nicolson scheme. The obtained set of linear equations is solved by the LDU decomposition method.

3. Numerical Experiments

3.1. Single Wave Propagation

Triggering the chemical solution at the edge of a petri dish generates a single chemical wave at the triggering point in a laboratory experiment. The chemical wave propagates across the dish. During the wave propagation, we observe flow velocity and chemical concentration at the
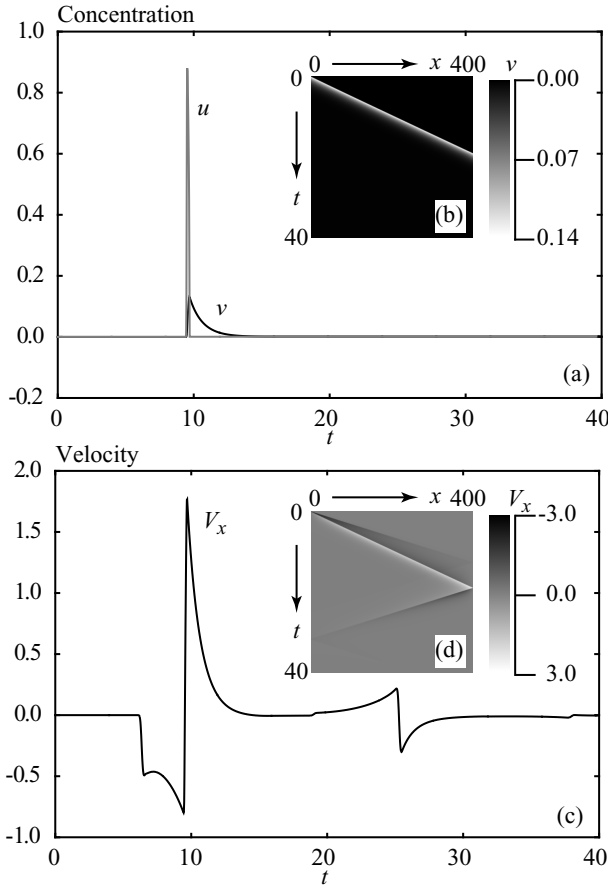
Figure 2: Numerical experiments for a single wave propagation. (a) Temporal changes of concentration distributions u and v at the center of the 1-dimensional domain and (b) spatio-temporal plot on v. (c) Temporal change of surface velocity V_x at the center of the 1-dimensional domain and (d) its spatio-temporal plot. In the spatio-temporal plots, gray level refers to the concentration of v in (b) and the surface flow velocity in (d). Parameter values utilized here were $\delta x = 1/5, \delta t = 1/1000, L_x = 400, D_u = 1.0, D_v = 0.6, p = 2.5, q = 5.0 \times 10^{-4}, \varepsilon = 0.01, D_d = 1000, a = 0.5, b = 500$. The single wave was generated at the the left side of the 1-dimensional domain at $t = 0$ by triggering u. Initial conditions for u, v, d were zero over the 1-dimensional domain.

center of the dish on the surface of the chemical solution. When the chemical wave reaches at the center of the dish, the surface flow velocity shows the maximum flow speed and alternates on its flow direction. The experimental result implies that the surface flow is caused by the concentration changes due to the chemical wave propagation. However, the experimental result also shows that the flow velocity on the surface at the center of the dish already exists before the chemical wave reaches there, as shown in Fig.4 of the reference [1]. Just after the triggering, the flow velocity at the center is increasing almost linearly, as time proceeds.

A numerical experiment was carried out to confirm the

validity of the proposed model when a single wave propagates. Figure 2 shows the result. The triggering time is $t = 0$. While the chemical wave reaches at the observation point of the center of the 1-dimensional domain at about $t = 10$, the absolute value of the surface flow velocity begins to increase at about $t = 7$. When the chemical wave reaches at the center, the flow velocity rapidly alternates on its direction at $t = 10$. Furthermore, at about $t = 15$ the flow velocity again alternates. This is caused by the reflection of the flow structure at the end of the domain [see the spatio-temporal image of Fig.2(d)]. These circumstances observed in the numerical experiment meet the results of laboratory experiments. However, the numerical experiment does not show the circumstance that the flow velocity begins to increase at the triggering time, as observed in the real laboratory experiments.

3.2. Wave Train Propagation

In real laboratory experiments, a chemical wave train also organizes a flow structure of the chemical solution. A core of a spiral pattern works as a pace-maker that produces a series of traveling chemical waves, namely, a chemical wave train. The two of authors (Miike and Sakurai) and their collaborators recently found an interesting flow fields. The novel points of the flow fields observed in their laboratory experiments are as follows [1]. Flow structures is organized along the horizontal (surface) direction and the structure travels also along the horizontal direction. The spatial wave length of the flow structure is much longer than that of the chemical wave train; the traveling speed of the flow structure is much higher than that of the chemical wave train. Flow direction at a particular point on the surface is alternating in time and space. The surface deformation in the depth direction is also organized.

We carried out a numerical experiment on a chemical wave train propagating in a 1-dimensional domain and a surface flow with the proposed model. To generate a wave train, we repeatedly triggered the center of the 1-dimensional domain at the time intervals of the oscillation period $\lambda = 2.0$. Figure 3(a) shows the spatial distributions of the chemical concentration v and the surface flow velocity V_x at $t = 195$. The spatial distribution on V_x shows the two surface flow structures, each of which has the pair of negative and positive flow velocity. The spatial wave length of the surface flow structure is much longer than that of a chemical wave. Since the surface velocity is caused by the spatial gradient of the chemical concentration, the precise wave length of the surface velocity is almost same to that of the chemical wave train. However, as time proceeds, the large structure of the surface flow is organized. Figure 3(b) shows the spatio-temporal plot of the propagating chemical wave train in the half of the 1-dimensional domain. The wave train propagates at the constant intervals. Figure 3(c) shows the spatio-temporal plot of the surface flow velocity. From the figures, we can recognize that the directions
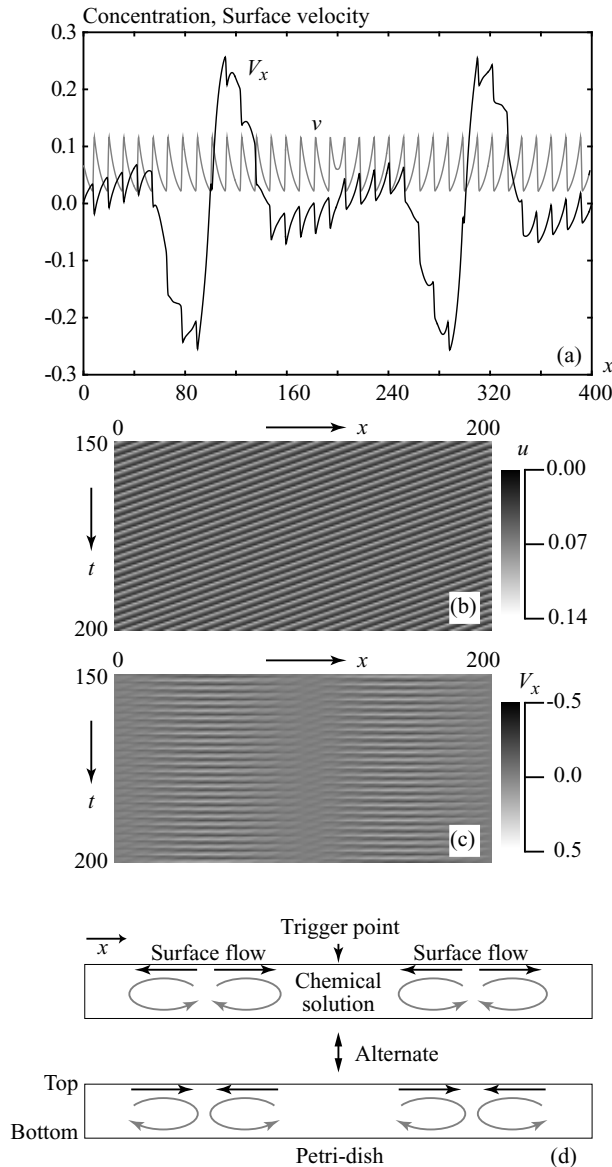
Figure 3: Numerical experiment for a wave train propagation. The center of the 1-dimensional domain was repeatedly triggered on u at the time intervals of $\lambda = 2.0$ for generating the wave train. (a) Spatial distributions of the concentration v and the surface flow velocity V_x at $t = 195$. Spatio-temporal plots of (b) the concentration v and (c) the surface flow velocity V_x in the ranges of $0 \leq x < 200$ and $150 \leq t < 200$. Parameter values utilized here were $\delta x = 1/5, \delta t = 1/1000, L_x = 400, D_u = 1.0, D_v = 0.6, p = 2.5, q = 5.0 \times 10^{-4}, \varepsilon = 0.01, D_d = 10000, a = 0.2, b = 1000$. Initial conditions for u, v, d were zero over the 1-dimensional domain. (d) Alternating global flow structures conjectured from (a) and (c).

of the flow structure changes alternatively as time proceeds and the flow structure propagates faster than the chemical wave train. Figure 3(d) shows the illustration of the 1-dimensional surface flow structure and the 2-dimensional one conjectured from the result of the numerical experiment of Figs. 3(a) and 3(c)

4. Conclusions

The present paper proposed a model that explains the phenomenon of the surface flow structure observed during a single wave or a wave train propagation in the Belousov-Zhabotinsky chemical reaction system. The proposed model utilizes the Oregonator model expressing the non-linear chemical reaction and the reaction-diffusion model expressing the spatially connected non-linear oscillators. The proposed model coupled the reaction-diffusion equation having the Oregonator model with the spring network model assuming the surface of the chemical solution. Through numerical experiments, we have confirmed that the proposed model can explain partially the characteristics observed in real laboratory experiments. However, we have still several points that we can not explain with the present proposed model. Furthermore, we have not shown that the surface deformation and 2- or 3-dimensional flow structure induced by traveling waves.

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