Self-maintained Movements of Droplets with Convection Flow

Hiroki Matsuno[1], Martin M. Hanczyc[2], and Takashi Ikegami[1]

[1] Department of General Systems Sciences, The Graduate School of Arts and Sciences, University of Tokyo, 3-8-1 Komaba, Tokyo 153-8902, Japan
hiroki@sacral.c.u-tokyo.ac.jp
[2] ProtoLife Srl, Via della Liberta 12, Marghera, Venezia 30175, Italy

Abstract. Running droplets have been studied recent years as dissipative macroscopic structures with locomotive capability, a characteristic of which is shared with biological systems. We constructed a numerical model of a droplet that integrates fluid dynamics and chemical reaction. Our results show that the chemical gradient generates droplet's motion, accompanied with convection flow. This convection flow contributes sustaining the chemical gradient, making a positive feedback loop. The simulated droplet self-maintains a chemical gradient, a prerequisite for locomotion, which constitutes a prototype of autonomous movement.

1 Introduction

Since the pioneering work of von Neumann [1] and succeeding studies by stimulated researchers, self-reproduction has been intensively studied in the field of artificial life [2, 3]. While self-reproduction is, beyond question, essential to life for its relevance to evolution, motility of biological systems is no less essential than self-reproduction.

Biological systems are non-equilibrium macroscopic structures, many of which show locomotive capability. As a dissipative structure, the natural cell receives resources from its environment and coverts them into waste through metabolism. The accumulated waste unless removed will saturate the local environment effectively slowing or stopping the metabolism that created it. Moving through the environment to obtain new resource allows biological systems to avoid the equilibrium. A self-movement becomes particularly important when a system acquires sensors and the adequate coupling between sensors and motors. A self-movement with sensors will differentiate context of the environment, which is food and where is enemy, to increase the value of survivability. We thus think the locomotion or exploratory behaviors is a basis of further evolution to take off.

In the field of artificial life, movement has drawn much attention through the recent enthusiasm for embodied cognition and situatedness. On the other hand there has not been much work to date on movement in simple chemically embodied systems such as protocells. Suzuki and Ikegami [4] have constructed an

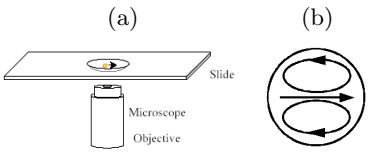

Fig. 1. (a) Experimental setup. The concave glass slide is filled with aqueous solution. An oil droplet is then added and observed by a microscope. (b) Schematic representation of the droplet with convection flow inside. This droplet moves to the right.

abstract model on a running cell in which metabolism and motility is coupled, employing a stochastic automata. Numerical result shows that the cell demonstrates the motility driven by inhomogeneous configurations of chemicals in the cell, which can be distinguished from Brownian motion.

From laboratory experiments it has been shown that oil droplets placed in aqueous media sometimes exhibit spontaneous movements. Difference in interfacial tension at the droplet boundary is responsible for the movement, known as Marangoni effect. For example, an oil droplet [5] is driven by receiving surfactants from the environment to create Marangoni effect, constituting a non-equilibrium structure with locomotive capability, the property of which is shared with living things, thus providing a prototype of biological movements. Recently we conducted a series of experiments on spontaneous motions of oil droplet with its boundary covered with fatty acid [Fig. 1 (a)] [6]. The oil droplet contains fatty acid anhydride that reacts with water at the boundary to produce fatty acid. Since the interfacial tension depends largely on the fatty acid concentration, inhomogeneity in the distribution of the chemical causes a Marangoni effect resulting in droplet's locomotion. Along with the movement, convection flow is observed as shown in Fig. 1 (b), whose axis coincides with the direction of movement.

Convection flow has been also observed in some of other studies but it remains unclear whether the convection flow is merely a byproduct of droplet motion or it contributes to the motion in a positive manner. Since in our experiment convection flow carries fatty acid anhydride toward the boundary or reaction region, it is implied that convection flow plays a role in enhancing locomotive behavior. It is then interesting in a sense that chemical gradient generates motion together with convection flow, which, in turn, sustains chemical gradient by providing reaction substrate, forming a circular relation and self-maintained movement. This paper aims to evaluate this conjecture. We have constructed a numerical model of a droplet that integrates fluid dynamics and chemical reaction. In the subsequent sections we explain our model in detail, report the result of droplet's motions and conclude with summary and discussion.

2 Numerical simulation

We introduce a model that qualitatively reproduces the behaviors observed in the experiment. As we saw, chemical reaction and convection flow are coupled and play a key role to sustain droplet motion. For decades, spatial structures formed through chemical reaction have been typically studied as reaction diffusion. To model running droplet we have to introduce the flow of the reaction field itself. Therefore, the model must include both fluid dynamics and a chemical reaction.

On the similar line Kitahata et al. [7] conducted numerical simulations which incorporate both convection flow and reaction diffusion system. In their model chemical gradient produced by BZ reaction generated the differentiated interfacial tension on the liquid-liquid interface. Assuming that the interface is a fixed straight line, it was shown that convection flow is generated near the interface due to Marangoni effect and moves along with chemical traveling wave. Our model differs in that it deals with the movement of droplet itself, which have the curved geometry and can change its shape. The governing equations are as follows.

Governing equations

$$\nabla \cdot \mathbf{u}(\mathbf{x},t) = 0 \tag{1}$$

$$(\frac{\partial}{\partial t} + \mathbf{u}(\mathbf{x},t)\cdot\nabla)\mathbf{u}(\mathbf{x},t) = -\frac{1}{\rho}\nabla P(\mathbf{x},t) + \nu\nabla^2\mathbf{u}(\mathbf{x},t) + aF_s\delta \tag{2}$$

$$F_s(\mathbf{x},t) = \gamma(v(\mathbf{x},t))\kappa\mathbf{n} + \nabla\gamma(v(\mathbf{x},t)) \tag{3}$$

$$\gamma(v(\mathbf{x},t)) = v(\mathbf{x},t) + b \tag{4}$$

$$(\frac{\partial}{\partial t} + \mathbf{u}(\mathbf{x},t)\cdot\nabla)v(\mathbf{x},t) = G(v(\mathbf{x},t))\delta + D_v\nabla^2 v(\mathbf{x},t) \tag{5}$$

$$G(v(\mathbf{x},t)) = \begin{cases} c, & \text{if } 0 \le x < 0.8 \\ 0.1c, & \text{else if } 0.8 \le x < 1 \\ 0, & \text{otherwise} \end{cases} \tag{6}$$

Equations (1)-(4) describe the dynamics of an incompressible fluid and (5) and (6) are for chemical reaction. Eq.(1) represents the conservation of mass. Eq.(2) represents the conservation of momentum or Navier-Stokes equation. The third term in the right hand side is a force at the interface which is defined in eq.(3). The first term in RHS of (3) constitutes interfacial tension. $\kappa, \mathbf{n}, \delta$ are curvature, normal vector of interface, delta function, respectively. Delta function takes a positive value at an interface , otherwise 0. γ is an intensity of interfacial tension which depends on chemical concentration. Here we simply assume it is linear to chemical concentration(4). The second term in eq.(3) is a force generated due to the difference of γ, that is, Marangoni effect. Eq.(5) is the same as reaction diffusion equation except for the advection term which enable the flow of chemical field. For simplicity, we deal with a single chemical species whose amount is represented with v. Chemical reaction is defined as in (6).

Numerical procedure

Space is two dimensional and described by a square mesh 64 by 64. We solve partial differential equations by finite difference method. Density function ϕ is defined to discriminate the droplet from the surrounding medium. It is set to 1 inside the droplet, otherwise 0 as an initial condition. ϕ is advected as (7). Interface between the two fluids can be detected by the difference in ϕ. δ function in eq.(2) and (5) can be substituted by $\nabla\phi$ (8). To obtain $\nabla\phi$, the stepwise-value ϕ needs to be smoothed out beforehand as numerical treatment. Here we apply eq.(10) to ϕ eight times, which gives about four mesh wide boundary. Two fluids can have different physical properties. When the densities of droplet fluid and surrounding media are ρ_1 and ρ_2 respectively, the density at the position $\mathbf{x}$ at time t is expressed as in (10). The kinematic viscocity is written similarly (11). Using these values, the governing equations can be solved as if we dealt with a single type of fluid. As a simulation scheme we used cubic-polynomial interpolation method. It interpolates the values between two neighboring mesh points using cubic polynomial to suppress numerical errors and is known for its simple algorithm [8]. Numerical procedure is summarized as follows.

$$(\frac{\partial}{\partial t} + \mathbf{u}(\mathbf{x},t)\cdot\nabla)\phi(\mathbf{x},t) = 0 \tag{7}$$

$$\delta = \nabla\phi \tag{8}$$

$$\begin{aligned}\phi_{i,j}^{new} = \frac{1}{2}\phi_{i,j} + \frac{1}{2}\cdot\frac{1}{1+4C_1+4C_2}\{&\phi_{i,j} + \\ &C_1(\phi_{i-1,j}+\phi_{i+1,j}+\phi_{i,j-1}+\phi_{i,j+1}) + \\ &C_2(\phi_{i-1,j-1}+\phi_{i-1,j+1}+\phi_{i+1,j-1}+\phi_{i+1,j+1})\} \\ &\left(C_1 = 1/(1+1/\sqrt{2}), C_2 = C_1/\sqrt{2}\right)\end{aligned} \tag{9}$$

$$\rho(\mathbf{x},t) = \phi(\mathbf{x},t)\rho_1 + (1-\phi(\mathbf{x},t))\rho_2 \tag{10}$$

$$\nu(\mathbf{x},t) = \phi(\mathbf{x},t)\nu_1 + (1-\phi(\mathbf{x},t))\nu_2 \tag{11}$$

1. set an initial condition as will be described in (12).
2. calculate the interfacial force F_s, using eq. (3).
3. – solve the Navier-Stokes equation (2) and continuity equation (1) to get u and P updated.
   – ϕ and v are also updated through advection and chemical reaction by eq. (7) and (5).
4. Iterate 2 and 3.

3 Results

In this section we report the simulation results of the above-mentioned model. We impose the chemical distribution with gradient as an initial condition. Chemical concentration is highest at left end of the droplet and is decreasing to the right

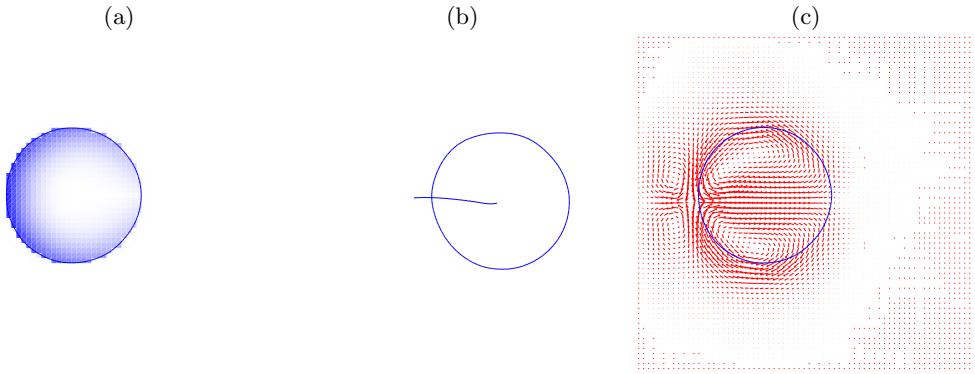

Fig. 2. (a)an initial condition: the chemical is asymmetrically distributed inside the droplet. The line showing the boundary of the droplet is drawn as the contour where $\phi = 0.5$. (b)The droplets marches from left to right. The line shows the trajectory of the center of mass. (c)The velocity field is shown. Each of short lines at mesh points represents a velocity vector at its location. Convection flow is observed whose axis is directed to the droplet's movement.

end. Besides, Chemical is set dense near droplet's boundary (Fig. 2 (a)). It is written as follows.

$$v = \begin{cases} |\theta|/\pi \cdot (|x|/R)^3, & \text{if } |x| \leq R \\ 0, & \text{otherwise} \end{cases} \tag{12}$$

Here, x, R, θ, are a positional vector, diameter of the droplet and radian measured from the center of droplet as the original. The parameters used in our simulations are listed at the end of this paper [table 1].

As the simulation proceeds, we can observe that the droplet moves to the right (Fig. 2 (b)). This rightward motion is produced because interfacial tension is stronger at the left boundary than at right one due to chemical gradient. During its locomotion velocity field is formed as shown in Fig. 2 (c). Inside the droplet we can see convection generated whose axis coincides with the direction in which droplet moves. The chemical flows within the droplet and then reaches the right side and then flows along the upper or lower boundary, gradually approaching the left side. This structure of convection flow can be equated with that observed in the experiments and reproduces it well.

As mentioned before, it is implied that chemical gradient is being sustained possibly because chemical reaction at an interface is balanced with convection flow which transports fresh chemical for the reaction. To verify this conjecture, we run simulations with the following two scenarios and compared them one another.

no reaction condition If there is no reaction at the interface, chemical gradient is expected to decline, flowing away from the boundary due to convection

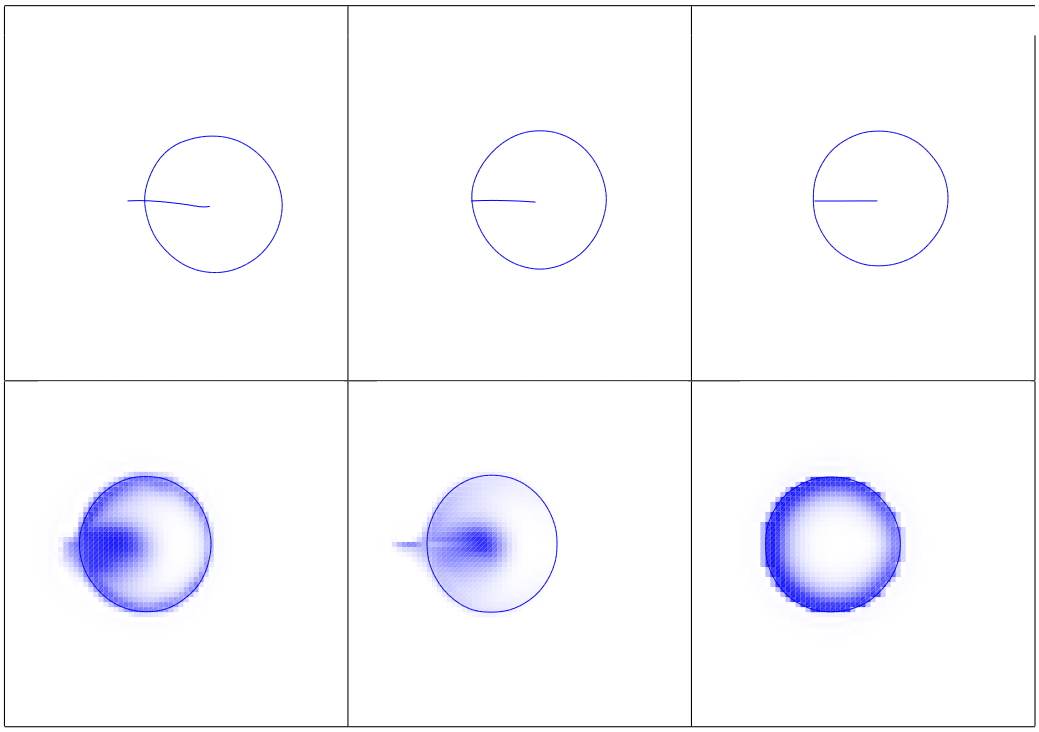

Fig. 3. (upper) Trajectory of droplets' movement. Each figure corresponds to a regular case (left), no chemical reaction case (middle) and no convection case (right). (lower) the snapshot of chemical gradient for each of three cases at the same time elapsed since the start.

flow. This condition is realized simply by ignoring the reaction term in Eq. 5.

no convection condition We conducted a simulation in which the initial chemical distribution is not altered by convection flow. The droplet's center of mass velocity $\int \phi \mathbf{u} dV / \int \phi dV$ (V: volume) is calculated from velocity field. Then we drift the droplet with this velocity uniformly. Chemical distribution moves in space without being deformed as if it were rigid body because its velocity gets independent from the position. While convection cannot affect the chemical gradient, chemical reaction can increase chemical product. Though this situation is physically impossible, it is conducted as virtual setting for comparison.

Results are shown in Fig. 3 and 4 along with the result of the regular case in which both reaction and convection are intact. Fig. 3 (upper) shows the distance of locomotion. In all cases droplet begins to travel and after a while cease to move. While the difference in the distance of displacement is not significant,

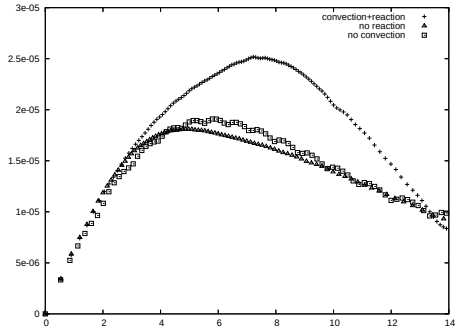

Fig. 4. The horizontal and vertical axes represent simulation time and the speed of droplet's center of mass, respectively. Regular case shows the highest speed among three scenarios.

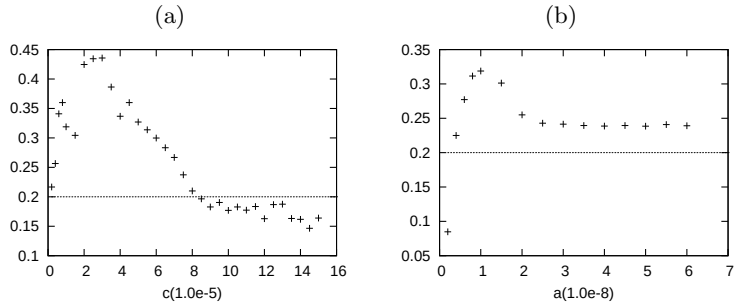

Fig. 5. The parameter dependencies on reaction rate c (a) and a, the coefficient of the force F_s (b). The horizontal and vertical axes stand for each of parameter values and the displacement distance, respectively. The horizontal dotted line represents the length of droplet's radius for comparison.

the droplet travels longer distance in the regular case than in the other two scenarios. Fig. 3 (lower) shows the snapshot of chemical gradient for each of three cases at the same time elapsed since the start. We can see that chemical gradient remains similar along the interface in the regular case. In no reaction case chemical is diluted and shows diminished gradient at the interface. On the contrary, chemicals are accumulated in no convection case, showing isotropic distribution. The reason of higher motility in the regular case is that chemical production is balanced with convection flow which takes away the chemical from the interface. Fig. 4 shows the speed of locomotion. The regular case demonstrates higher speed than the other two. These results indicate that droplet can have higher locomotive capability when convection and reaction works cooperatively.

To understand further the relation between the chemical reaction and convection flow, we tested how the droplet's behavior depends on the reaction rate

c and the parameter a which determines the convection flow. Fig. 5 (a) shows dependencies on c with a fixed. We can see that around a specific parameter region the droplet accomplishes the maximum displacement distance. Fig. 5 (b) illustrates dependencies on c. Again, specific c region is favorable for locomotion. Both reaction rate and Marangoni effect have an influence on motility.

4 Conclusion and future work

We qualitatively reproduced the movement of a droplet, using the model which couples the fluid dynamics and chemical reaction. Staring from the initial condition in which chemical gradient is appropriately set inside the droplet, we observed that droplet shows directional motion. This movement is generated by the imbalance in interfacial tension which can be locally different, depending on the chemical concentration on the interface. Along with its motion, convection flow is also observed inside the droplet, whose axis coincides with the direction in which the droplet marches. The capability of movement is influenced by the two parameters, intensity of interfacial tension depending on the chemical and reaction rate of chemical at the interface. We checked that the maximum traveling distance is achieved at the parameter region where the two parameters take specific values to balance the two contrary effects: chemical reaction which increases the chemical concentration till the saturation and convection flow that carries away chemicals from the interface. Lack of either one would cause faster decline of chemical gradient, resulting in poor motility. Besides, to see whether the convection flow contributes to the droplet's motion in a positive manner, we compare the observed running behavior with that in a virtual scenario in which convection flow doesn't affect the initial distribution of the chemical. While the droplet exhibits directional movement in both cases, it displays higher motility when convection flow is incorporated. Therefore we can say that there exists a case where convection flow enhances the running behavior.

Chemical gradient is a prerequisite for generating motion. In addition, the chemical gradient is being sustained and generated by the droplet itself in the course of time. Decades ago, autopoiesis was suggested as biological model which sustains itself by reproducing its components and boundary conditions. In our model the chemical gradient and convection flow constitute a positive feedback loop which sustain the droplet's motion. This is a self-sustenance of motion, as autopoiesis is for reproduction, and can be regarded as extension to a more general conception where the motion is also self-maintained as reproduction is.

A special emphasis has been put on self-reproduction in the field of artificial life both from theoretical and empirical points of views [2, 9–13], while movements have not drawn much attention until recent years. However, when it comes to autonomy, the essential characteristics in which life manifests itself, we can hardly imagine life without movement.

In biological experiments chemotaxis of bacteria and other organisms has been studied, which is considered to be the prototype of cognitive behaviors [14]. From very different points of view in psychology, Gibson[15] and more recently

parameter	description	value
ρ_1	density (droplet)	1.2
ρ_2	density (surrounding media)	1.0
ν_1	kinematic viscosity (droplet)	1.7×10^{-6}
ν_2	kinematic viscosity (surrounding media)	1.0×10^{-6}
R	the radius of a droplet	0.2
δh	mesh size	1.56×10^{-2}
Re	Reynolds number	15
D_v	chemical diffusion coefficient	0.2
a	the coefficient of force F_s	1.0×10^{-8}
b	the constant in the interfacial tension term	0.2
c	chemical reation rate	1.0×10^{-5}

Table 1. Parameters: Values above are used in our simulation, unless specified in the text. To determine Reynolds number by $Re = uL/\nu$, the diameter of the droplet $2R$ and the average velocity at the center of droplet's mass are used as representative length L and velocity u, respectively.

O'Regan and Noë [16] in particular contributed much to the shift of paradigms in cognition from a passive view that sensor input is a signal fed from environments to agents to an alternative view that input is generated by agents through their exploratory movements.

Adaptive behavior and cognitive capabilities are now considered in terms of correlation between movements and sensor input. Various computational models and robotic experiments have been suggested to illuminate sensor-motor loops [17]. Our research on the droplet serves as a model which exemplifies the self-maintenance of movement and bridges the gap between protocell models and locomotive agent models, which have been sometimes considered unrelated to one another.

Some issues remain to be addressed as future work. Spatio-temporal structures formed by reaction diffusion system can be introduced in our model. An interplay between diffusion and convection might have some significance. In laboratory experiment Kitahata [7], for example, showed that droplets loaded with BZ reaction diffusion system demonstrate oscillatory movements driven by oscillatory chemical patterns. Besides, it is widely reported that convection and diffusion take place inside the cell. Our model can be used to simulate and analyze such situations. Secondly, we imposed a chemical gradient as an initial condition to initiate movement. On the other hand, droplets in the experiment spontaneously give rise to symmetry break without special treatment of chemical gradient as an initial condition. Symmetry breaking and initiation of movement are left to future work. Thirdly, The shape of the object, in general, can affect the mode of its movement or be altered along with the movement. For example, camphor, which is also known to show running behavior, generates directional or circular motion, depending on its shape. Relation between the shape and its mode of motion can also be of importance.

Acknowledgements

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