

Chemical Basis for Minimal Cognition

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Abstract We have developed a simple chemical system capable of self-movement in order to study the physicochemical origins of movement. We propose how this system may be useful in the study of minimal perception and cognition. The system consists simply of an oil droplet in an aqueous environment. A chemical reaction within the oil droplet induces an instability, the symmetry of the oil droplet breaks, and the droplet begins to move through the aqueous phase. The complement of physical phenomena that is then generated indicates the presence of feedback cycles that, as will be argued, form the basis for self-regulation, homeostasis, and perhaps an extended form of autopoiesis. We discuss the result that simple chemical systems are capable of sensory-motor coupling and possess a homeodynamic state from which cognitive processes may emerge.

Keywords

Chemotaxis, cognition, homeostasis, interfacial tension, oil droplet

1 Introduction

Perception, intelligence, and higher-order cognitive processes as currently understood are rooted in sensory-motor coupling in organisms [1, 2, 7, 15, 31, 45]. Indeed, this coupling is being designed into robotic systems to create artificial “living” machines (EcoBot [27] and SlugBot [22]). However, the fundamental basis for cognition may already be present in simple nonliving physical systems that possess a limited suite of properties also found in living systems. Intelligence could then be traced down to physicochemical phenomena, such as thermodynamic fluctuations in open nonequilibrium systems of rather simple composition that form by self-assembly, as we will see in our chemical systems. So the question is, how can we derive sophisticated intelligence from a merely thermodynamic system? In other words, when is it necessary to use the *intentional stance* [12] to describe a system’s behavior—for example, by using *sensing*, or *cognition*, instead of reaction, or hysteresis?

Over the past few years we have been developing self-assembling chemical systems that are capable of motility [17, 35, 36]. The chemical systems consist of an oil droplet in an aqueous water phase. The aqueous phase contains a surfactant that forms the interface between the water and the oil and modulates the interfacial tension between the droplet of oil and its environment. We embed a chemical precursor (oleic anhydride) in the oil phase (nitrobenzene) that hydrolyzes into more surfactant when it comes in contact with the water phase at the oil-water interface. This reaction not only powers the droplet to move in the aqueous phase, but also allows for sustained movement as long as enough precursor oil remains in the droplet.

By embedding a catabolic chemical reaction in a self-assembled oil droplet body, we have determined some of the conditions necessary to establish an interactive loop that involves the global movement of the

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system (and thus can be interpreted as sensory-motor coupling) in a wet chemistry model defined by only five chemical components, including water. In this article we describe the phenomena involved, in particular the mechanism of motility, and we speculate on how such a bottom-up approach can define the naturally robust architecture necessary to evolve minimal cognition on top of an elementary type of sensory-motor coupling. By way of studying this system, we reorganize the concept of autopoiesis, the self-regulatory mechanism of a cell, around the terms of motility and homeodynamics. These are the vocabularies proposed for considering a system as a cognitive system rather than solely a chemical system.

2 Description of the System

We study the movement of an oil droplet in an aqueous phase optically. The diameter of the droplet can range from a few hundred microns to a few centimeters. An oil phase introduced into an aqueous phase self-assembles into an oil droplet. Without any chemical or physical perturbation, the oil droplet will not move. It is only by creating an instability in the system that the oil droplet becomes dynamic. The instability in our system arises from the hydrolysis of the oleic anhydride in the oil phase, and within seconds after the introduction of the oil to the water phase, the droplet starts moving (see the Appendix on materials and methods).

The movement of the droplet is accompanied by convective flow inside the droplet. The convective flow is established in the following steps. First, a chemical reaction occurs on the surface of a droplet. As more products are accumulated, the local pH decreases. The reaction does not occur evenly along the surface, so that the local pH varies. We have measured the interfacial tension as a function of the surfactant to show that the surface tension increases when the pH decreases (see Figure 2 in Section 3.1). Therefore the local interfacial tension also varies along the surface. The subsequent interfacial tension gradient defines the Marangoni force, whose direction is tangential to the surface. The Marangoni force drives the local flow along the surface (called the Marangoni flow). A Marangoni instability describes the flow of surfactant (liquid) along an interface to equilibrate an imbalance in interfacial tension and is the underlying physical principle behind the “tears of wine” effect [43]. Once the initial symmetry breaks by fluctuation, a pair of convective flows is organized. With numerical simulation [25], we demonstrated that the initial symmetry breakdown is followed by quadratic-vortex formation, and pair convective flow follows.

The convective flow structure in the droplet brings fresh precursor to one pole of the droplet while controlling the release of products on the opposite pole. Experimentally, the convective flow is observed inside the moving oil droplet, with the centerline of flow along the direction of droplet movement (see Figure 1). We believe that convective flow serves to create key feedback cycles in our system, and due to convection, the reaction and movement will be sustained.

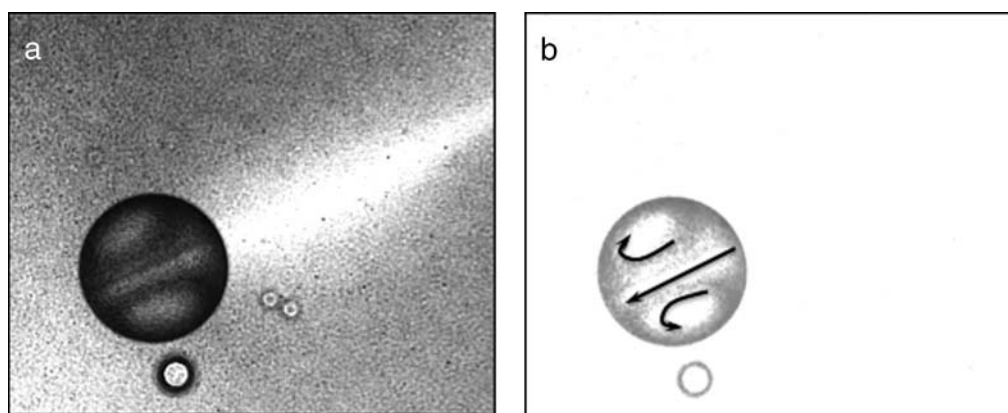


Figure 1. DIC micrograph of a self-moving oil droplet with internal convection. (a) The droplet is moving toward the bottom left while creating a bright trail. The characteristic flow pattern associated with convection is clearly seen within the oil droplet. (b) Overlay of flow patterns seen in the oil droplet. The diameter of the droplet is nearly 0.1 mm.

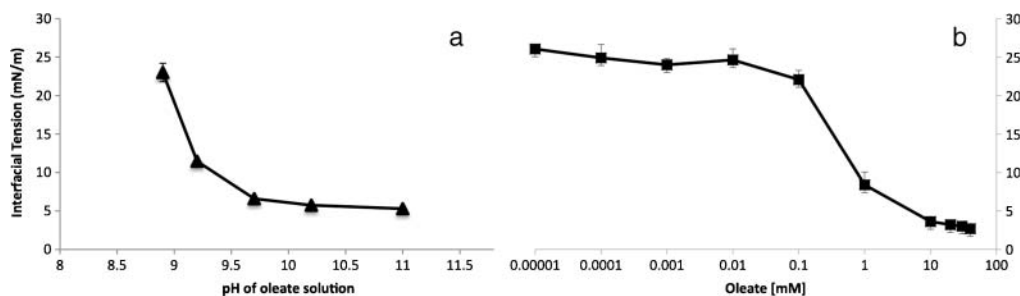


Figure 2. Change in interfacial tension of a nitrobenzene oil droplet measured by pendant drop tensiometry and profile analysis. (a) The interfacial tension of a pure nitrobenzene droplet in a 10 mM oleate solution with varying pH. (b) The interfacial tension of a pure nitrobenzene droplet in solution at pH 11 with varying oleate concentration. Each measurement was taken in triplicate. The interfacial tension of a drop of nitrobenzene at pH 11 with no oleate present is 27 mN/m.

The mechanism of self-motion is complex even in this simple system, with the law of reciprocal action likely being dominant in the early stages and hydrodynamic pressure due to the fluid dynamics in the latter stages of movement. Both the convective flow and the direction of movement are governed by unequal interfacial tension at the oil droplet boundary. The sustained imbalance in interfacial tension as the droplet moves is likely caused by an observed local pH gradient that surrounds the droplet. This chemical gradient is created by the droplet itself as a product of the chemical reaction. The self-generated gradient can be overridden by an externally imposed pH gradient, and therefore the direction of droplet motion may also be controlled. We observe that the droplet “senses” the gradient in the environment (either self-generated or externally imposed) because the internal flow patterns and direction of droplet movement change accordingly. When the droplet moves predictably within a pH gradient, it exhibits chemotaxis in its most basic sense: directional movement governed by an external chemical gradient. (See [17] for a complete chemical description of the system.)

3 Observations and Discussions

3.1 Sensory-Motor Coupling

Here we explain in more detail how a sensory-motor coupling arises in a simple chemical system. All the complexity of this self-moving motion ultimately depends on the oil-water interface and the integration of the sensor and actuator into the same dynamic structure. The interface is not a hard-shell container, but a soft and flexible boundary under tension that can interact physically and chemically with the local environment. It is the interface that is observationally responsive when exogenous chemical gradients are added to the system. Therefore, we consider the interface to be the sensor for the system.

The motor for the system arises from the flow structures observed within the oil droplet. When the interface senses a chemical gradient, such as in pH, an imbalance in the tension surrounding the droplet results in flow structures due to a Marangoni instability. The observed flow structures with a droplet, such as convection (see Figure 1), can act as the motor in the system.

We argue that both sensor and motor are present once the oil self-assembles into a droplet, and if so, we can then begin to test the system for sensory-motor coupling. As the droplet ages, the anhydride precursor fuel is hydrolyzed at the oil-water interface to produce more oleate and protons. Both products can affect the tension at the interface, as shown in Figure 2, and these factors modulate the responsiveness of the sensor. The oil droplets are usually added to an aqueous phase containing 10 mM oleate at pH 11. The production of more oleate surfactant has little effect on the tension surrounding the droplet, as the tension is already quite low (Figure 2b). However, a local decrease in pH can have a large effect, quickly reaching a tension maximum at pH 9 (Figure 2a). It is noted in experiments with pH-sensitive dyes that the pH locally can decrease by several units, as low as pH 7 [17]. This change in pH therefore can have a large effect on the tension surrounding the droplet and can cause a Marangoni instability and induce flow. The sensing of local changes in pH by the interface induces flow, and the

droplet starts to move. This form of pH-directed chemotaxis, as seen in our system, would be an example of a type of sensory-motor coupling. We note that the droplets move chemotactically in pH gradients even when the precursor fuel is not added to the system, but the movement is not sustained and will stop once it successfully equilibrates any imbalance in tension (typically in a few seconds). This illustrates that instabilities in self-assembled systems induced externally can be resolved quickly. Instead, what we show here is that instabilities generated from within the system itself may allow for sustained movement. The sustainability associated with this system is its novel feature. The system becomes sustainable by circulating the reactants and products effectively as organized by the convective flow.

There are several mechanisms for chemotactic behavior in unicellular living systems. The mechanisms for the motor include the rotation of the flagella in prokaryotes (e.g., *E. coli* [23]), actin-myosin dynamics of the cytoskeleton (e.g., *Dictyostelium* [6, 21, 37, 47]), movement of the cilia (in eukaryotes such as *Tetrahymena* [10]), and gliding in cyanobacteria [18]. Each of these motility mechanisms is linked to specific transmembrane receptors on the cell, acting as sensors. The sensors relay information about the extracellular environment to the mechanical motors through intracellular signaling pathways involving numerous proteins and small molecules [30, 39]. Although varying in sophistication [16, 46], these sensory-motor couplings allow the organism to perform chemotaxis to find nutrients and avoid waste and other poisons. The effects of sensing can be complex, with factors such as sustainability and long- and short-term memory coming into play. There have been some simulation studies that qualitatively reproduce the motion of the amoeba as well as chemotaxis [4, 29], where different diffusion rates and production rates assigned for actin and myosin are responsible for the amoeba's motility. The mechanism of movement and sensory-motor coupling in oil droplets is much simpler and does not break down into modular units. In a sense, the sensor and the motor are integrated into one, and one does not make sense without the other. This kind of simple chemotaxis mechanism, as found in our oil droplets, is not known in extant living systems.

Studying sensory-motor coupling with a simple chemical system has advantages. We do not need to design and manufacture special devices or organs for sensor and actuator functions. Instead, the system self-organizes, and as a result many such oil droplets can be made simply and economically. This makes our chemical system attractive for those interested in studying self-organized systems that possess sensory-motor coupling, lifelike behaviors (e.g., chemotaxis), and possibly the basic elements of computing. The oil droplet system, because of its simplicity in composition, dynamic behavior in multi-dimensional spaces, and possible emergent behaviors, could be used as an artificial life model system in a chemistry laboratory, just as the game of Life is used in the virtual laboratory.

3.2 Shape Matters

By responding to a pH gradient with concomitant convective flow and movement, the droplet behaves as if it can “perceive” the environment. (When and in what sense we can use this term “perception” will be discussed in the following subsections.) We believe that the geometry of the interface shape can control sensitivity to the environment. Also, geometry-induced fluctuations can be the source of fluctuation in motion [24, 28, 34, 38]. A coupling between fluctuations in interface geometry and fluctuations in motion may be linked with the idea of biological autonomy. For example, it has been found that by mechanically pushing the cytoplasm of a cell (e.g., *Dictyostelium*) one can elicit directional locomotion [11]. The asymmetrical change of a boundary shape causes a polarization in actin and myosin protein filaments, causing directional motion [44]. This internal polarization of biological chemicals may be related to our observations. In our typical experiments with droplets about 100 μm or less in diameter, we do not observe any fluctuations in droplet shape as the droplet moves. However, in larger droplets up to 0.5 cm in diameter, fluctuations in shape become readily apparent, as shown in the examples in Figure 3 (see also [32]). In such examples, both the distortion in shape and the fluctuation in motion (velocity, direction) vary on the time scale of seconds (Figure 4). The larger droplets are more easily deformable, especially in the presence of surfactants, where the forces of gravity and perhaps flow structures counteract the Laplace pressure, which maintains the spherical shape [14]. In addition, increasing the droplet size enhances the instability of the internal flow, as the Reynolds number is proportional to

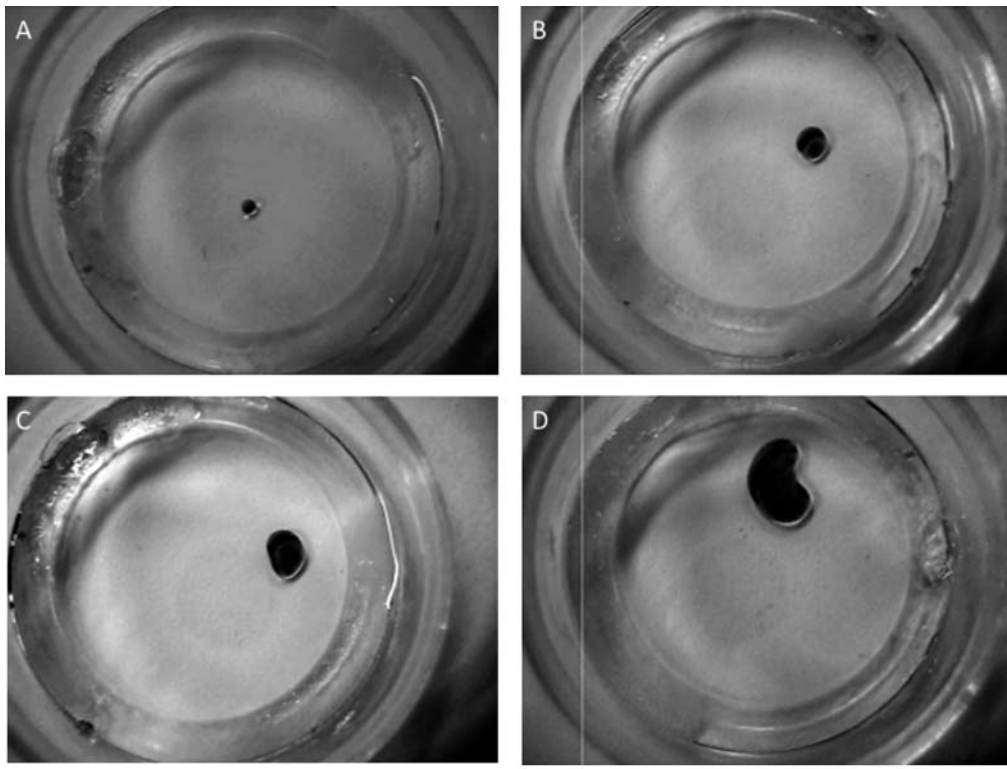


Figure 3. Shape change in the boundary structure in moving droplets of different size. Droplets of size 1, 5, 10, and 30 μL were added to the same aqueous phase and analyzed for fluctuations in geometry and movement (panels A, B, C, D, respectively). Size bar, 1 cm.

the size. Figure 3 shows the transition from the spherical shape to a horseshoe shape. We note that straight directional motion is most supported by the horseshoe shape. This distorted boundary shape may not support the convection flow seen in the smaller droplets. Therefore, the mechanism of self-movement shifts from convection-driven to shape-driven above around a few hundred microns in droplet size. The detailed analysis of this phenomenon will be reported elsewhere [19].

3.3 Autopoiesis and Autonomy

Behavior can be treated as an extension of a basic idea of biological autonomy called autopoiesis [40]. Autopoiesis is a self-regulating mechanism of an internal metabolic network that maintains the boundary of the cell. The autopoietic cell can be explicitly modeled by a simple stochastic automaton on a

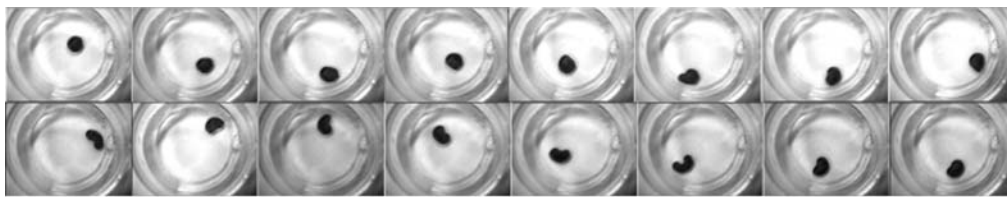


Figure 4. Fluctuations in droplet shape and movement over time. One droplet of 30- μL volume was added to the aqueous phase, and each frame was taken at 8-s intervals. Size bar, 1 cm.

two-dimensional grid with abstract chemistry [42]. In this model, abstract chemistry is used to exemplify the concept. Substrates of the environment are transcribed into membrane materials by a catalytic particle. Those membrane particles will enclose the catalytic particle, so that when a membrane is broken, it is immediately repaired by the catalytic particle; see Figure 5. This self-consistent relationship realizes the concept of autopoiesis.

Varela and Maturana [26, 40–42] argued for organization of a minimal living system in terms of autopoiesis. They discussed that a transition from a physicochemical system to a minimal living system happens when a system performs an action on “what is missing on the part of the system,” in Varela’s terminology. For instance, a system repairs its physicochemical boundary to maintain its “identity” (as in our chemical system and Varela’s original experiment and model). This compensating process allows for the emergence of a cognitive unit on top of the physicochemical layer. Also, Varela argued that the self-regularity, as well as the environmental regularities, is the result of coevolution between living systems and their environment. Namely, the environment is also a part of living organisms. This is exactly what we observed in our chemical experiments. We tend to think that the droplet becomes autonomous by its own mechanism, but this is not completely accurate. Its environmental factors (pH and oleate concentration, both of which are influenced by the action of the droplet; see [17]) and also temporal treatment (the freshness of the neat oil phase, pipetting actions) make autonomous movement possible. Even after the autonomous droplet emerges, it is still controlled by the environment and its own temporal changes. This is what we consider to be the congruent regularity of the droplet motion, which is the product of both droplet and environment.

Here we explore how autopoiesis is further developed through self-motility. The original concept of autopoiesis does not explicitly assume the phenomenon of self-motion, nor does it argue what kinds of self-motion are effective; but our chemical experiments and some simulated models (e.g., [33, 34]) exemplify the case where an artificial organism maintains the autopoietic system by the emergence of the motile state. We consider the coexistence of both spontaneous and reflective (chemotactic) behavior an extension of autopoiesis. That is, depending on the internal state and the environmental condition, a droplet moves around in a certain way, at the same time changing the environmental condition (i.e., the modified environment can function as an external memory reservoir). Speculatively, a droplet may be able to “select” the action between autonomous and reflective to compensate for changes in the velocity of the reaction. In addition, there are some other types of action selection where a droplet

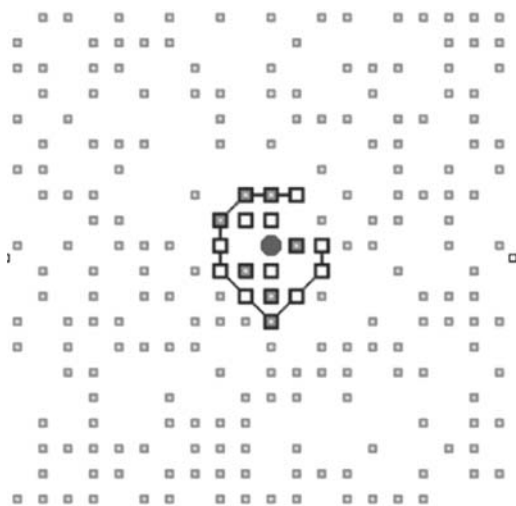


Figure 5. The original simulation model of an autopoietic cell [26]. The many small squares are substrate (S) particles. The larger squares, sometimes connected with other squares, are the link (L) particles. The single circular pattern in the middle is the catalytic (C) particle.

spontaneously changes its direction of motion. For example, by changing the strengths of outgoing flow of product from the interface (through the law of reciprocal action), the droplet can navigate. The potential reorganization of the droplet internal state as it switches between action modes is currently under investigation.

The above dynamic component extends the notion of autopoiesis in that we can interpret the droplet's motion as action selection. Based on observation of the present droplet experiment, together with the previous studies on autopoietic cells (e.g., [33, 34]), we have a revised notion of autopoiesis as follows.

1. In order to have an autopoietic system, a boundary is a necessary ingredient that separates the inside from the outside. The self-consistent relationship between the internal chemical network and the organization of the boundary is what we call the autopoietic property of the unit. (This is the original definition of autopoiesis.)
2. What is missing in the original picture of autopoiesis is *temporal organization*. The above formal definition of autopoiesis does not tell whether autopoiesis is indeed realizable in living systems. We need to know the dynamic stability and durability of autopoietic properties in the real world. In the current oil droplet experiment, the droplet moves, sometimes demonstrating chaotic itinerancy, and by doing so acquires autopoiesis. In other words, self-motility prevents the system from relaxing the nonequilibrium state, so that it can sustain autopoietic properties over time.
3. Dynamic autopoiesis adds some new properties to a system. First, shape fluctuation of the boundary assumes new degrees of freedom for the autopoietic cell. Whether the shape is relatively round or rugged changes the compensating (therefore "intentional") action pattern of the cell. Second, the dynamic component lengthens the life span of the autopoietic cell (in the previous models, the life span, defined by the time during which a droplet moves, of the autopoietic cell was not controlled [34]). In other words, the dynamic autopoiesis changes the time structure of the system.

By considering the dynamic nature of autopoiesis, we noticed that a static boundary could be replaced by a convective flow structure. Now we have to answer the rather provocative challenge by one of the reviewers: Why do we need to distinguish between sensing and reacting? This question naturally leads to the question of when and where we switch the vocabulary from reaction to sensing and/or perception, and also, why we use the word sensory-motor coupling instead of (for example) climbing down the free-energy landscape. One answer is proposed by taking into account Varela's work, the emergence of compensating action to maintain the individuality of a droplet. Let us argue this in terms of the game of Life, where we note several similarities to the chemical model described here.

In the game of Life, recurring patterns are termed *gliders*, and we use that term to describe the entire dynamics instead of using the underlying bit patterns. Using that vocabulary can help to compress the game of Life phenomena. Our first motivation was to find a gliderlike object in the chemical system, and we have identified the droplet as such an object. Once the moving droplets appear, it is more efficient to describe the phenomena in terms of droplets, because the reaction system and convective flow patterns are hidden behind the outward phenomena. This is also true for describing robots' behavior. Their movements are nothing more than a pattern of the electric circuit and a sensory pattern hidden behind it.

When an object emerges, the underlying constituents become unseen, and we can switch our way of description. Of course, in this droplet experiment, we see a clear connection between the underlying chemical layer and the droplet layer, so that switching vocabulary is not strictly necessary. However, when we have successive hierarchical layers (e.g., functional differentiation of the droplets) with outward function overshadowing the lower-layer dynamics, we have more reason to use psychological words in describing our system and developing our understanding of it. This droplet experiment is one such step toward the complete vocabulary switch. Further, the droplet holds its own individuality, namely, a droplet becomes robust against perturbation, which is different from a glider. Therefore, as

a physicochemical entity, the droplet becomes an individual unit. In the next subsection, we consider the underlying mechanism that holds individuality, namely, homeodynamics.

3.4 From Homeostasis to Homeodynamics

Any perturbations in the interfacial tension of the boundary structure tend to be balanced through the flow of surfactant. Once the tension forces around the droplet are balanced, the droplet stops moving. However, the emergent convective flow structure brings fresh precursor to the surface, where it becomes metabolized, and the system therefore maintains an imbalance in tension allowing for sustained movement. This is an example of the homeodynamic state. Originally, homeostasis was described as a property of a self-regulating system that sustains critical variables in a certain range [5, 9]. In the case of homeodynamics, a system changes the parameters or boundary condition to adapt to the environment. In doing so, the system dynamically organizes its own parameters. This leads to a peculiar state that we call the homeodynamic state.

This idea of varying the system's parameters is based on Ashby's ultrastability [1]. In contrast with the simple feedback loop that tunes the sensory-motor system, a second feedback loop exists to tune the critical system parameters. The difference between variables and parameters is made explicit when writing down equations. Variables evolve temporally, and parameter values are fixed in time. But in real systems, the difference is likely not so simplistic. In case of the oil droplet, variables could be the center of mass of the droplet, the velocity of the center mass, the amounts of chemicals, and so on. The parameters could be droplet size, pH, viscosity, reaction rate, and so on. But, as we have seen so far, those variables and parameters are mutually dependent. The critical parameters are changed to break up the homeostatic state to make a system more robust against the environment, yielding a homeodynamic state.

Ashby proposed the concept of ultrastability for designing a brain system. We propose that a self-moving oil droplet is a physical realization of homeodynamics and thus present it as a minimal cognitive model. Self-movement regulates the chemical reaction. Also, when a droplet is perturbed externally, it responds by resetting the flow pattern or changing the boundary shape. Shapes and flow structures are the essential parameters of the droplet. The viable constraints on the droplet are determined by those essential variables, so that positive feedback from the convection flow to the chemical reaction is the second feedback in terms of Ashby's ultrastability.

In recent studies of autonomous robotics, cognitive behaviors are characterized by a sensory-motor coupling that can also be termed *embodiment*. The advantage of embodiment has been repeatedly stressed in the field of robotics for the last two decades [3, 8, 31]. A missing notion in the robotics field is the self-organization of self-movement and homeodynamics (a notable exception is Di Paolo's study of homeoadaptation with an autonomous robot [13]). By pushing a step forward with the homeodynamic aspect underlying any cognitive behavior, we say that the droplet can be a critical example for studying minimal cognition. Indeed, the transition from homeostatic self (self-maintained statically) to homeodynamic self (self-sustained dynamically) [20] emphasizes the potential for homeostasis as a source for purposeful behavior even in simple systems.

4 Conclusion

Even simple chemical systems may tell us something about complex emergent phenomena such as cognition. Using a bottom-up approach, we produced a simple oil droplet capable of sensing and modifying its environment, which results in autonomous self-movement of the droplet through an aqueous phase. The boundary at the liquid-liquid interface serves as a highly sensitive and dynamic structure that can perceive the environment. Once a pH gradient in the environment surrounding the oil droplet is perceived, the droplet responds with movement within the gradient. The embedded chemistry of the system fuels and reinforces sustained movement of the droplet. In this way the droplet maintains itself through homeodynamic processes. We then begin to see an extended view of the self and autopoiesis as a structure that maintains itself and its boundary through physically dynamic processes such as movement. Such systems have a more active communion with their environments through perception,

decision-making, and even cognition. We hope to understand the fundamental aspects of cognition through the intersection of simple physicochemical systems and cognitive science. Different from the mere physicochemical process, a living system preserves its own identity and consistency with respect to the environment by constantly readjusting the homeostatic state. Sensory-motor coupling, rooted in homeodynamics, is the key to understanding minimal cognition and physical intelligence conscripted and exploited by living systems [20].

When do we need to take the “intentional stance” [12] and resort to a new vocabulary? A critical point is when a system sustains its structure by homeodynamics, that is, a cyclical relationship between sustaining its internal process and self-motility, and thus behaviors emerge that can be named and interpreted. Once self-motility becomes inevitable, we begin to see an extended view of the self; autopoiesis is no longer just a stationary state that maintains itself. By realizing the homeodynamic dimension of autopoiesis, we speculate that there would be no life without self-movement.

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Appendix: Materials and Methods

A.1 Oleate Surfactant

Neat oleic acid oil (NuChek Prep, Elysian, MN) was added to alkaline water to make varying concentrations of oleate micelles at pH 11. NaOH was adjusted to make oleate solutions at varying pH for tensiometry.

A.2 Oil phase

Neat oil of oleate anhydride (Fluka) was added to nitrobenzene (Fluka) at 0.5 M. Pure anhydride was used in neat oil form. Anhydride oil stocks were stored away from direct light and under argon.

A.3 Glass Slide Experiment with DIC Microscopy

Surfactant (100 mL) was added to glass slides with a concave depression (VWR, Milan, Italy) of 1.5-cm diameter. An oil droplet was then added, and movement was monitored using microscopy. Running droplets were analyzed in real time using an inverted Olympus differential interference contrast (DIC) microscope, BX51, equipped with a Toshiba CCD camera connected to a video-recording system (Sony, WV-DR9).

A.4 Glass Dish Experiment

Aqueous phase (800 mL) was added to a glass dish of 35-mm diameter with a 27-mm-diameter quartz base. An oil droplet was then added, and movement was recorded using an iSight digital video camera and iMovie software.

A.5 Tensiometry

Interfacial tension of the oil-water interface was determined using a PAT1D tensiometer (Sinterface) by the pendent drop method using the Sinterface software. All samples were prepared with nitrobenzene as the internal phase and an aqueous phase containing varying concentrations of oleate or 10 mM oleate with varying pH. All values were taken after the tension reached a steady state, and each condition was tested in triplicate.

