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 physico-chemical origins of movement, perception and cognition. The system consists simply of an oil droplet in an aqueous environment. A chemical reaction embedded in the oil droplet induces 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 argues for the presence of feedback cycles that form the basis for self-regulation, homeostasis, and autopoiesis. We argue here that simple chemical systems are capable of sensory-motor coupling and possess a homeodynamic state from which cognitive processes may emerge.

Keywords:

Chemotaxis, oil droplet, interfacial tension, cognition, homostasis
Introduction

It is believed that perception, intelligence and higher order cognitive processes start at the level of sensory-motor coupling in organisms\textsuperscript{12,34,5}. Indeed this coupling is being designed into robotic systems to create artificial ‘living’ machines (i.e. EcoBot\textsuperscript{6} and SlugBot\textsuperscript{7}). However, the fundamental basis for cognition may already be present in simple non-living physical systems that possess a limited suite of life-like properties. It may be that intelligence can be traced to physical phenomena such as thermodynamic fluctuations in open systems of even simplistic composition that form by self-assembly. Over the past few years we have been developing self-assembling chemical systems that are capable of motility\textsuperscript{9,10}. The chemical system consists 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 some precursor oil remains in the droplet.

By embedding a catalytic chemical reaction in a self-assembled oil droplet body, we have determined some of the conditions necessary to establish sensory-motor coupling in a wet chemistry model defined by only five chemical components including water. In this paper we describe the phenomena of the system including the mechanism of movement and we speculate on how such a bottom-up approach can define the architecture necessary to evolve intelligence in more complex living systems.

Description of the system

When oil is introduced to the aqueous phase, it self assembles into an oil droplet. Without the chemical reaction, the hydrolysis of oleic anhydride, the oil droplet will not move. It is only by creating an instability in the system that the oil droplet becomes dynamic. The instability on our system arises due to the presence 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. The initial breakage of symmetry initiates a convective flow structure in the droplet which then brings fresh precursor to one pole of the droplet while controlling the release of products on the opposite pole. We note that convective flow is generated inside the moving oil droplet, with the centerline of flow commensurate with the direction of droplet movement (see Figure 1). The emergence of convective flow was also observed by simulating the fluid dynamics integrated with chemical reactions (Matsuno et al., 2007, ACAL 07, Springer, p.179, Springer). We believe that the convective flow serves to create key feedback cycles in our system, and due to the convection, the reaction and movement will be sustained.
The mechanism of self-motion is complex even in this simple system with the law reciprocal action likely being dominant in the early stages and hydrodynamic pressure due to the fluid dynamics in the latter stages of movement. The convective flow and direction of movement are governed by unequal interfacial tension at the oil droplet boundary. This imbalance in the interfacial tension is likely caused by an observed local pH gradient that surrounds the droplet. This chemical gradient is created by the droplet itself. 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 can observe that the droplet senses the gradient in the environment (either self-generated or externally imposed) by changing its internal flow patterns. When the droplet moves predictably within a pH gradient, it exhibits chemotaxis (see Hanczyc et al, 2007 for a complete chemical description of the system).

**Sensory-motor coupling**

All the complexity of this self-moving motion ultimately comes from the oil-water interface. It is not a hard shell container but it is a soft and flexible boundary under tension that interacts with the local environment. An imbalance in the tension surrounding the droplet results in flow structures due to a Marangoni instability. Flow structures such as convection (see Figure 1) can act as the motor in the system. Chemotaxis, as seen in our system, is a simple example of such sensory-motor coupling. How interfacial tension changes within the varying parameters of our system is illustrated in Figure 2. As the system proceeds, the anhydride precursor is hydrolyzed at the interface to produce more oleate and protons. Both products can affect the tension at the interface as shown in Figure 2. The oil droplets are usually added to an aqueous phase containing 10mM oleate at pH 11. The production of more oleate surfactant has little affect 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 (Hanczyc et al, 2007). 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. 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 the movement 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 may allow for sustained, self-regulated dynamics such as self-movement.

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 motor functions. Instead, the system self-organizes and as a result many such oil droplets can be made simply and cheaply. This makes our chemical system attractive for those interested in studying self-organized systems that possess sensory-motor coupling and life-like behaviors. The oil droplet system because of
its simplicity in composition, dynamic behavior in multidimensional spaces, and 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.

**Shape matters**

By responding to a pH gradient with concomitant convective flow and movement, the droplet behaves as if it can perceive the environment. 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. 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. The asymmetrical change of a boundary shape causes a polarization in actin and myosin protein filaments, causing directional motion. This internal polarization of biological chemicals may be related to our observations. In our typical experiments with droplets of about 100 micron 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 ref). 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, which decrease the Laplace pressure. In addition increasing the droplet size enhances the instability of the internal flow, as the Reynolds number is proportional to the size. When we increase the size of the droplet from 100 um to a few cm in diameter, the shape of droplet changes drastically. Figure 3 shows the transition from a spherical shape to the boomerang shape. We note that straight directional motion is most supported by the boomerang shape.

This action selection behavior is similar to what we see in simulation where a two dimensional organism moves in order to maintain its boundary structure. This continual maintenance of the boundary structure by the emergence of the motile state can be considered to be a form of autopoiesis. Autopoiesis is a self-regulating mechanism of an internal metabolic network that maintains the boundary of the cell. This autopoietic cell can be modeled by a simple stochastic automaton with abstract chemistry. Three chemical species (C, S, L) are prepared. C catalyzes to form L from 2 S. L is bonding together to make a membrane loop and once this loop encloses C, the loop becomes autopoietic. Because by the enclosure, the C cannot escape the loop, so that enough amount of L is stocked within a cell which enables continuous repairing of the cell membrane (Varela et al. 1974).

A drawback of this first model is that the cell only moves randomly and breaks up quickly. We thus modified the system by adding some special rules to organize self-movement. Our second model organism can now move by continuously self-repairing the membrane, but it failed to show any gradient-climbing behavior.
(climbing the gradient of S particles) (Figure 5). This may be due to the fact that the autopoietic cell can only survive in the narrow range of environments that support a certain substrate density. In the third model, we impose that the loop membrane structure never breaks up but the length can become longer or shorter. This case is most similar to our laboratory oil droplet system where the membrane interface always exists but the shape can change allowing for modulation of surface area. Our finding with this third model is that the cell with a round shape can move around and show gradient-climbing behavior. But a more deformed cell (which as a result has a larger surface area) fails to show the chemotactic behavior. Compared with the autopoietic toy cell models above, our oil droplets correspond well to the round cell in the third model. They respond to both internal and external perturbations with movement in order to maintain their boundary structures.

How shape changes alter the flow patterns of the droplets is now under examination. As we understand, modern biological cells utilize shapes rather than convection flow to organize their motion, but at the same time we know that there exist flow of molecules inside a cell. Therefore the trade-off between shapes and flow is an interesting feature to examine in both living and model systems.

**Autonomy**

The oil droplets move because they seek to maintain their boundary structure while their embedded metabolism continuously supplies an instability at the interface. An externally imposed instability can be sensed by the droplet resulting in chemotaxis, highlighting the droplets sensitivity to environmental perturbations. Remodeling of the pH landscape by the droplet itself is responsible for the droplet’s autonomous motion. The coexistence of both autonomous and chemotactic behavior is where we find the autonomy in this droplet. Depending on the internal state and the environmental condition, droplets may be able to “select” the action. The potential reorganization of the droplet internal state as it switches between autonomous and chemotactic mode is currently under investigation.

**Homeodynamics**

The droplet seeks to balance any perturbations in the interfacial tension of the boundary structure through the flow of surfactant. Once the tension forces around the droplet are balanced the droplet stops moving. However the emergent convective flow structure beings fresh precursor to the surface where it becomes metabolized and the system therefore maintains the imbalance in tension allowing for sustained movement. This is an example of the homeodynamic state. The original homeostasis is a property of a self-regulating system that sustains critical variables in a certain range. In case of homeodynamics, a system changes the parameters or boundary condition to adapt to the environment. In doing so, a
system dynamically organizes parameters of the system, we name it homeodynamics.

This idea of varying the system’s meta-parameter is based on R. Ashby’s ultra-stability (1960). Assume that we have a set of chemicals. Those chemicals can constitute a dynamic state that can be made comparable to a self-preserving attractor, which we like to label as a primitive form of self. But these self-preserving attractors can exist only under an adequate parameter set that controls their stability. Thus, the parameter range becomes a viability constraint because out of this range, self-preserving property scarcely appears.

This idea of viability constraint is the core idea of R. Ashby’s ultra-stability, where he illustrated double feedback interactions between a system and an environment. A primary feedback loop is a mutual interaction between complex sensory and motor channels and the environment. Another feedback goes from viability constraints to a reacting part through essential variables that control the reacting part.

Usually, the second feedback loop is executed only intermittently because it changes the meta-parameters of the system so that it drastically changes the behavior. When the parameter values are out of the viability constraints, the second feedback adjusts the essential parameters to let the system move towards the attractors (stationary dynamic states).

The difference between variables and parameters are made explicit when writing down equations. Variables temporally evolve, and parameter values are fixed in time. But in real systems, the difference is likely not so simple. In case of oil droplet, variables are the center mass of the droplet, velocity of the center mass, the amount of chemicals, etc. The parameters are droplet size, pH, viscosity, reaction rate, etc. But as we have seen so far, those variables and parameters are mutually dependent.

We propose that self-moving oil droplet is a natural realization of ultra-stability. Self-movement regulates the chemical reaction and when a droplet is perturbed externally, a droplet responds to by resetting the flow pattern or changing the boundary shape. Shapes and flow structures are the essential parameters to the droplet. The viable constraints of 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 ultra-stability.

Ashby proposed the concept of ultra-stability for designing cognitive behaviors. In recent studies of autonomous robotics, cognitive behaviors are characterized by the sensory-motor coupling (that can also be termed embodiment). Advantage of the embodiment has been repeatedly stressed in the field of robotics (Braitenberg 1984, Brooks 1999) for last two decades. A missing part in 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 (2000)). Therefore, 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)\textsuperscript{17} emphasizes the potentiality of homeostasis even in simple systems as a source for purposeful behavior.

\textbf{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 that 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 imbedded chemistry of the system fuels and reinforces sustained movement of the droplet. In this way the droplet is trying to maintain 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 physico-chemical systems and cognitive science. Different from the mere physico-chemical process, a living system preserves its own identity and consistency with respect to the environment. This homeostasis, rooted in sensory-motor coupling, is the key to understanding minimal cognition and physical intelligence conscripted and exploited by living systems (Ikegami, T. et al., 2008, BioSys., 91, p.388].

Acknowledgements

Methods
Figure 1. DIC micrograph of a self-moving oil droplet with internal convection. A) The droplet is moving towards 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 size of the droplet is nearly 0.1mm in diameter.

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 10mM oleate solution with varying pH. B) The interfacial tension of a pure nitrobenzene droplet in a 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 27mN/m.
Figure 3. Shape change in the boundary structure in moving droplets of different size. Droplets of different size (1, 5, 10, and 30 ul) were added to the same aqueous phase and analyzed for fluctuations in geometry and movement (panels A, B, C, D respectively). Size bar, 1cm.

Figure 4. Fluctuations in droplet shape and movement over time. One droplet of 30ul volume was added to the aqueous phase, and each frame was taken at 8 second intervals. Size bar, 1cm.
Figure 5: A simulation of an autopoietic cell system on a two dimensional plane over time. Different symbols denote different particle types and the cell is defined as a unit surrounded by the linked particles (Suzuki, K and Ikegami, T., 2004).

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