Dynamic Self-organization in an Open Reaction Network as a Fundamental Mechanism for the Emergence of Life

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Abstract

The emergence of life on the earth has attracted intense attention but still remained an unsolved question. A key problem is that it has been left unclear why a living organism can have self-organizing ability leading to highly ordered structures and evolutionary behavior. This work reveals by computer simulation and experiments that a stationary state of an open reaction network, into which some source substances flow at constant rates, really has such self-organizing ability. The point is that reaction and diffusion processes in an open reaction network are irreversible and always forced to approach equilibrium. Therefore, they necessarily reach a stationary state in which they approach equilibrium to the largest extent as a whole and attain a full balance. This means that a stationary state of an open reaction network is firmly stabilized by irreversible reaction and diffusion processes and kept stable against fluctuation, namely it has ability to organize itself. A stationary state of an open reaction network is also flexible in structure and can evolve based on its own self-organizing ability through interaction with the environment. Thus, this work provides a new general mechanism of self-organization and evolution in a prebiotic chemical system, which is expected to have acted as a fundamental principle for the emergence of life on the earth. It is interesting to note that a network of reversible processes in a machine has no self-organizing ability because a reversible process has no property of spontaneously and irreversibly happening in a particular direction.

Introduction

The emergence of life on the earth is one of the biggest topics in recent scientific research. A large number of studies has been made on relevant chemical substances and organization and many interesting findings have been reported.[1-6] However, it has still been left unclear how living organisms have produced highly organized structures composed of complex biopolymers such as enzymes, RNA, and DNA. For solving this problem, it is essential to reveal how a prebiotic chemical system can have self-organizing ability leading to such highly ordered structures.

It is said that not a small number of researchers assert that the emergence of life on the primitive earth happened by lucky accidents under favorable conditions,[1,4,7] but such an idea looks too optimistic. It cannot give a meaningful probability of the emergence of life because it needs repeated occurrence of lucky accidents until the emergence of the first

living organism. Moreover, this idea cannot explain the emergence of free spirit in living things because a reaction system investigated under this idea is in an entirely passive state against an attack of the external world and has no ability to organize itself.

Thus, it is indispensable to clarify how a prebiotic chemical system gained self-organizing ability, as mentioned above. Several ideas or approaches have been proposed about this issue. The formation of dynamic order such as chemical oscillations and spatiotemporal patterns, called dissipative structures or self-organization by Prigogine,[8] has attracted intense attention because of their similarity to nervous impulses in animals and patterns on bodies of zebras, leopards, and tropical fishes.[8,9] However, phenomena studied till now are still far in quality from self-organization in living organisms. In addition, such dynamic order arises from particular chemical reactions involving autocatalytic processes and lacks sufficient flexibility for evolution through interaction with the environment. Self-organization in a reaction system has also been studied in complexity science. Kauffman and his group reported by computer simulation that a rection network comes to have collective catalytic actions when it gets sufficiently complex and such a reaction network has ability to replicate itself.[1] For explaining the freedom of mind in living things from inevitable natural laws, workers in complexity science also reported that life emerges at the edge of chaos.[1,10] However, these are only theoretical models. In fact, Mitchell pointed out that it has remained an unsolved question how theoretical models in complexity science are realized based on physicochemical laws of real substances.[11] On the other hand, Luisi and coworkers reported, according to the idea of autopoiesis system,[12] a simple reaction system using a reverse micelle, which mimics chemical processes in a biological cell.[4,13] However, this system is still mainly formed by static self-assembly due to intermolecular interactions. Thus, no convincing mechanism for explaining dynamic self-organization in living things has been reported to date.

As a new attempt to deal with the above issue, the present work has investigated whether a stationary state of an open reaction network, hereafter abbreviated to ORN, has self-organizing ability or not. An ORN is regarded as the best model of the primitive life and thus such study should be of great importance but strangely, little attention has been paid to it. This is most probably because a stationary state of an ORN resembles a static equilibrium state and has appeared to involve no significant mechanism leading to dynamic self-organization. We have found that reaction and diffusion processes have a unique property of spontaneously and irreversibly changing in a particular direction and thus a network of them gives rise to dynamic self-organization. Another possible reason for little attention to a stationary state of an ORN is that it is a complex system with no definite boundary and has not been easy to deal with both theoretically and experimentally. We have succeeded in surmounting such difficulty by using computer simulation and experiments using an electrochemical system.

Results
1. Computer Simulation

Recent advancement of geophysics has disclosed a promising possibility that the first living organism emerged near the bottom of a small long pore in a mineral (FeS) precipitate in the vicinity of a hydrothermal vent in a primitive deep sea.[14,15] It is expected that various chemical substances discharged from a hydrothermal vent diffused into such a small pore and reacted with each other and thus formed an ORN. This expectation is

reasonable because reaction products with relatively large sizes and hence small diffusion constants will have accumulated within a small long pore. Namely, an ORN can be formed by differences in diffusion constants between entering chemical (source) substances and reaction products.

Figure 1 shows a model of such an ORN, in which only chemical substances of meaningful concentrations are picked out, others being neglected. Circles stand for chemical substances, different numerals referring to different chemical substances. Thus, a numeral i refers to a chemical substance R_i. A pair of arrows refers to a chemical reaction. Accordingly, Fig. 1 shows that R_1 and R_2 react and produce R_7 and R_8, and so on. All reactions are assumed to be elementary reactions and thus their rates, $\mathcal{R}_r$, can be expressed by second-order rate equations. For example, the rate of the reaction of R_1 and R_2 is given, by using the rate of change in the concentration of R_1, as follows.

$$-dC_1/dt = k_f C_1 C_2 - k_b C_7 C_8, \tag{1}$$

Inflow: $R_1 \sim R_6$ and R_{13}

Outflow: (a) $R_{43} \sim R_{49}$ or (b) all

Fig. 1. A model of an open reaction network (ORN) used for computer simulation.

where C_i is the concentration of R_i and k_f and k_b are the rate constants of the forward and the backward processes of reaction, respectively. In Fig. 1, chemical substances, R_1 to R_6 and R_{13} flow into an ORN, i.e. these are source substances, while all or some of reaction products are discharged from an ORN. The rate of the inflow or outflow of R_i, $\mathcal{R}_{di}$, can be approximately expressed, based on Fick's law of diffusion, as follows.

$$dC_i^{in}/dt = k_{di}\left(C_i^{ex} - C_i^{in}\right) \tag{2}$$

where k_{di} is the rate constant for the inflow or outflow of R_i and C_i^{ex} and C_i^{in} are the concentrations of R_i in the exterior and in the interior of an ORN, respectively. (The superscript "in" for C_i^{in} is omitted in case no confusion happens.) The diffusion rate for organic molecules in an aqueous solution is in general much higher than the reaction rate and we in the present work assume that chemical substances in an ORN have a spatially uniform distribution. Calculations were done by using an ODE (ordinary differential equation) solver in MATLAB software (MathWorks).

Figure 2 shows time courses of concentrations C_i in an ORN in two cases: (a) Only final reaction products, R_{43} to R_{49}, are discharged from an ORN, and (b) all reaction products, R_7 to R_{12} and R_{14} to R_{49}, are discharged. Reaction conditions are summarized as follows: the external concentrations C_i^{ex} of source substances are (2~8) $\times 10^{-3}$ M (M = mol dm^{-3}), those of discharged reaction products are 5×10^{-4} M for all, the initial concentrations of chemical substances in an ORN are 1×10^{-3} M for all, k_f is 0.6~1.1 M^{-1} s^{-1}, and k_f/k_b is 1.4~2.6, in both (a) and (b). On the other hand, k_{di} is $(0.94~1.0)\times10^{-4}$ s^{-1} in (a) and $(0.94~1.0)\times10^{-5}$ s^{-1} in (b).

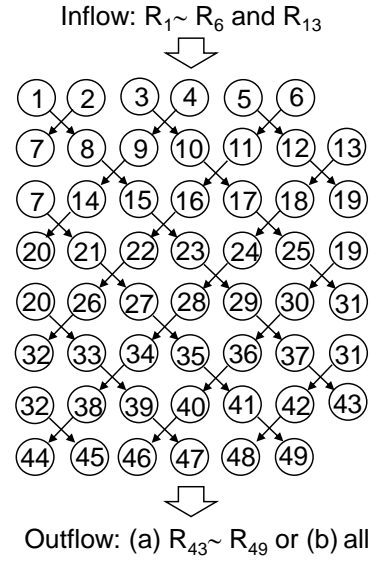The results of Fig. 2 indicate that the concentrations C_i of chemical substances R_i in an ORN initially change in complex ways but gradually approach constant and finally become constant, indicating that an ORN lastly reaches a stationary state. Calculations were done under various reaction conditions. A stationary state was achieved in all cases. The concentrations of chemical substances in a stationary state increased nearly in parallel to uniform increases in the external concentrations of source substances. On the other hand, the concentration profile in a stationary state little depended on such uniform increases in the external concentrations of source substances, though it strongly depended on their non-uniform changes. Similarly, the concentration profile little depended on uniform increases in k_f and k_{di} though it depended on their non-uniform changes. Interestingly, the concentration profile was kept the same when the initial concentrations of almost all chemical substances in an ORN were chosen to be zero, with the other reaction conditions kept unchanged, indicating that source substances mainly determine what ORN is formed.

Fig. 2. Time courses of the concentrations C_i of chemical substances R_i in an ORN in two cases: (a) only R_{43} to R_{49} are discharged and (b) all, i.e. R_7 to R_{12} and R_{14} to R_{49} are discharged.

Calculations were also done for some ORN's having different structures from Fig. 1, and we confirmed that essentially the same results as Fig. 2 were obtained. Furthermore, we investigated the behavior of ORN's in which all reactions are catalyzed by chemical substances within them. The catalytic action was introduced by expressing the rate constants k_f and k_b for catalyzed reactions in the form of $k_f = k_f{'} C_k$ and $k_b = k_b{'} C_k$ where C_k is the concentration of R_k acting as a catalyst and $k_f{'}$ and $k_b{'}$ are constant. Again, essentially the same results as Fig. 2 were obtained when $k_f{'} C_k$ and $k_b{'} C_k$ were chosen to be about 10^4 or 10^8 times as large as k_f and k_b for non-catalyzed reactions in Fig. 2.

Prigogine reported that the entropy production rate in a non-equilibrium open system, in which various flow processes in the near-equilibrium linear region are coupled with each other, some of the flow processes being placed under constant thermodynamic forces, takes a minimum in a stationary state.[9] Therefore, we investigated how the entropy production rate σ in an ORN changes when it approaches a stationary state. The σ is given in the form of the product of the rate of each flow and the thermodynamic force for it and can be expressed as follows.[9]
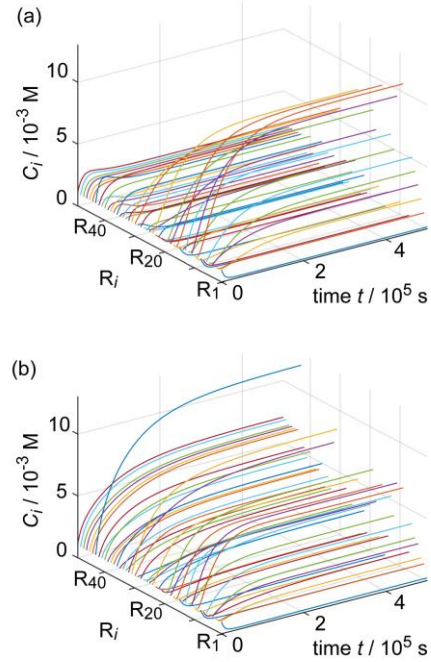
$$\sigma = \sigma_1 + \sigma_2 \tag{3}$$

$$\sigma_1 = \sum_i k_{di} V\left(C_i^{ex} - C_i^{in}\right)\left(\mu_i^{ex} - \mu_i^{in}\right) / T \tag{3a}$$

$$\sigma_2 = \sum_j \upsilon_j\left(A_j/T\right) \tag{3b}$$

where σ_1 refers to the entropy production rate by the inflows or outflows of chemical substances R_i while σ_2 does that by chemical reactions in an ORN. In Eq. (3a), μ_i^{ex} and μ_i^{in} are the chemical potentials of R_i in the exterior and in the interior of an ORN, respectively. V is the volume of an ORN and T the temperature. Thus, $k_{di} V\left(C_i^{ex} - C_i^{in}\right)$ refers to the rate of the inflow or outflow of R_i and $\left(\mu_i^{ex} - \mu_i^{in}\right) / T$ does the thermodynamic force for it. On the other hand, in Eq. (3b), υ_j expresses the rate of the j-th reaction, defined by

$$\upsilon_j = d\xi_j/dt = d(n_{pj}/v_{pj})/dt \tag{3c}$$

where ξ_j is the degree of extent of reaction, dn_{pj} a change in the amount of substance for a chemical substance R_p by the j-th reaction, and v_{pj} the stoichiometric coefficient for R_p in the j-th reaction (v_{pj} is defined as negative for reactants and positive for products, note also that n_{pj}/v_{pj} is independent of p). According to this definition, $-dC_1/dt$ in Eq. (1) is expressed as υ/V. The quantity A_j in Eq. (3b) is the affinity of the j-th reaction, given as $A_j = -\sum_p v_{pj}\mu_p$.[9] Thus, υ_j refers to the rate of the j-th reaction, as mentioned above, and $\left(A_j/T\right)$ does the thermodynamic force for it. The rate υ_j can be expressed by using A_j as follows,[9]

$$\upsilon_j = \upsilon_{jf}\left[1 - \exp\left(-A_j/RT\right)\right] \tag{3d}$$

where υ_{jf} is the rate of the forward process of the j-th reaction. Equation (3d) indicates that υ_j is in proportion to the thermodynamic force $\left(A_j/T\right)$ when $|A_j/RT| \ll 1$, namely the j-th reaction is in the linear region under this condition.

Figure 3 shows time courses of the entropy-production rates, σ_1, σ_2 and σ, for an ORN of Fig. 1 at 273.15K. The results (a) and (b) are obtained under the same reaction conditions as (a) and (b) of Fig. 2, respectively. In case (a), σ_1 initially increases and then decreases and becomes constant, while σ_2 starts from a high value and decreases, then increases and becomes constant. Thus, the sum of them, $\sigma = \sigma_1 + \sigma_2$, monotonously decreases and becomes constant. This result can be explained as follows. The reaction rates at the initial stage are slightly higher than the rates of inflows of source substances in case (a). Therefore, reactions in an ORN are initially dominant and thus σ_2 starts from a high value and decreases because of decreases in C_i in an ORN. Then, inflows of source substances follow such reactions, accompanied by an increase in σ_1. The
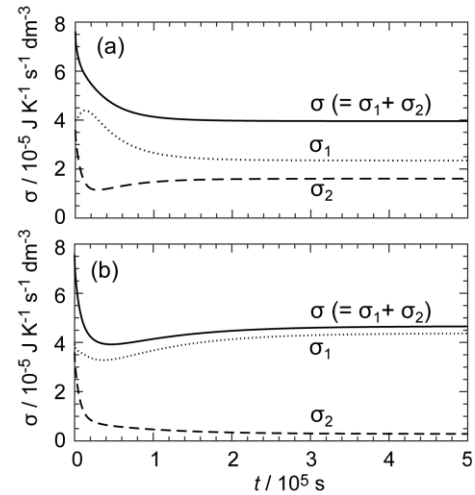

Fig. 3. Time courses of the entropy production rate, σ_1, σ_2, and σ, in an ORN of Fig. 1 at 273.15K. The above results (a) and (b) are obtained under the same reaction conditions as (a) and (b) of Fig. 2, respectively.

rates of inflows of source substances soon reach maxima and decrease because the concentrations of source substances in the interior of an ORN increase and come close to those in the exterior. Thus, σ_1 also reaches a peak and decreases. Finally, increases in the concentrations of chemical substances in an ORN by continuous inflows of source substances lead to an increase in the number of chemical reactions and hence an increase in σ_2. The result in case (b) can be explained in a similar way. In this way, time courses of σ_1, σ_2 and σ teach us how an ORN reaches a stationary state.

Now, the time courses of the entropy production rates, σ_1, σ_2 and σ, in Fig. 3 show that they become constant after the lapse of a sufficiently long time both in case (a) and (b), as mentioned above. The attainment of constant σ_1, σ_2 and σ means the achievement of constant reaction and diffusion rates, i.e. the attainment of a stationary state. Essentially the same results were obtained for σ_1, σ_2 and σ under other reaction conditions and for ORN's having different structures from Fig. 1. Note here that the σ ($= \sigma_1 + \sigma_2$) in Fig. 3(a) indeed takes a minimum in a stationary state, in agreement with Prigogine's theorem mentioned above, but the σ in Fig. 3(b) does not, namely it takes a minimum before an ORN reaches a stationary state. The result of Fig. 3(b) is not special but similar behavior is often observed under other reaction conditions. A plausible reason for such a discrepancy is that most of individual reactions in ORN's under the reaction conditions in the present work are in the nonlinear region and do not meet the necessary conditions for Prigogine's theorem. In addition, the mode of reaction coupling in an ORN in case (b) is complex and it is unclear which reactions or which course of reactions is placed under a constant thermodynamic force. Anyway, the result of Fig. 3(b) shows that we cannot necessarily say that the σ in an ORN monotonously decreases when it approaches a stationary state.

2. Experimental Investigation

Properties of an ORN were also investigated by experiments. We used an electrochemical system such as illustrated in Fig. 4, in which two Pt electrodes (anode AN and cathode CA) and five Pt probes, made by using photolithography, are immersed in the acidic aqueous electrolyte containing redox couples. The application of a voltage between anode and cathode causes an electric current to flow in the redox electrolyte, and five Pt probes measure the redox potential of the redox electrolyte at each position because a Pt probe and the redox electrolyte are in electron-transfer equilibrium. The electric current in the redox electrolyte flows by an electron-transfer reaction (or electron hopping) between a reductant R and an oxidant Ox in the electrolyte as well as the diffusion of them. As explained later, the former process makes a main contribution to the electric current. Therefore, when the
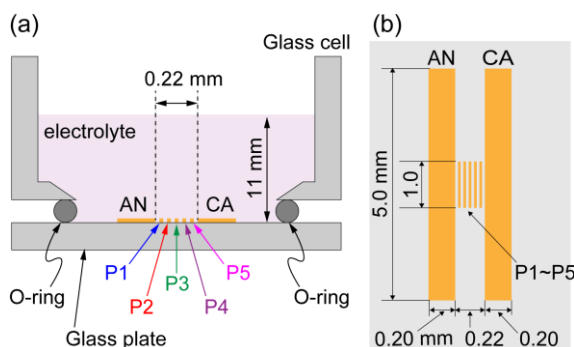

Fig. 4. (a) A cross-sectional view of an electrochemical cell and (b) a top view of two Pt electrodes (anode AN and cathode CA) and five Pt probes (P1, P2, P3, P4, and P5).

redox electrolyte contains a variety of redox couples, the electric current flows by consecutive occurrences of electron-transfer reactions of a type, $R_i + Ox_j \rightleftharpoons Ox_i + R_j$, where the subscripts i and j refer to the kind of redox couples. Thus, the redox couples in the redox electrolyte form an ORN in which source substances are supplied by electrode reactions at the anode and some reaction products are discharged at electrode reactions at the cathode, both at constant rates. We can then expect the achievement of a stationary state in this electrochemical system.

Figure 5 shows time courses of (a) the electric current I and (b) the potentials U of five Pt probes, simultaneously measured after the application of a potential of 200 mV between anode and cathode. The redox

Fig. 5. Time courses of (a) the electric current I and (b) the potentials U of five Pt probes vs. CA (cathode), simultaneously measured after the application of a potential of 200 mV between anode and cathode.

electrolyte contained 0.1 M H_2SO_4 and three redox couples of Fe^{2+}/Fe^{3+} (0.77 V), $Fe(bpy)_3^{2+}/Fe(bpy)_3^{3+}$ (1.03 V), and $Fe(phen)_3^{2+}/Fe(phen)_3^{3+}$ (1.12 V), where bpy stands for bipyridyl and phen does o-phenanthroline, and numerals in parentheses refer to the standard electrode potentials. The concentration of every reductant R_i and oxidant Ox_i was chosen to be 0.1 M at the stage of no electron transfer reaction between redox couples. The electric current initially decayed and became constant, indicating that a stationary state was formed. The potentials of five Pt probes, which measured the redox potential of the redox electrolyte at each position, as mentioned above, also initially changed and became constant with roughly the same potential difference from each other, again indicating that a stationary state was formed. These results can be explained as follows. With no potential application between anode and cathode, the redox electrolyte is in equilibrium and the concentrations of R_i and Ox_i are each homogeneous throughout the electrolyte. Under the application of a potential, the concentrations of R_i and Ox_i at the anode and cathode surfaces are determined by the applied potential. In a stationary state, the electric current density does not change along a line of electric current. Thus, the redox potential in the redox electrolyte and the concentrations of R_i and Ox_i both change linearly with a spatial distance along a line of electric current. This explains why the potentials of five Pt probes became constant with roughly the same potential difference in a stationary state.

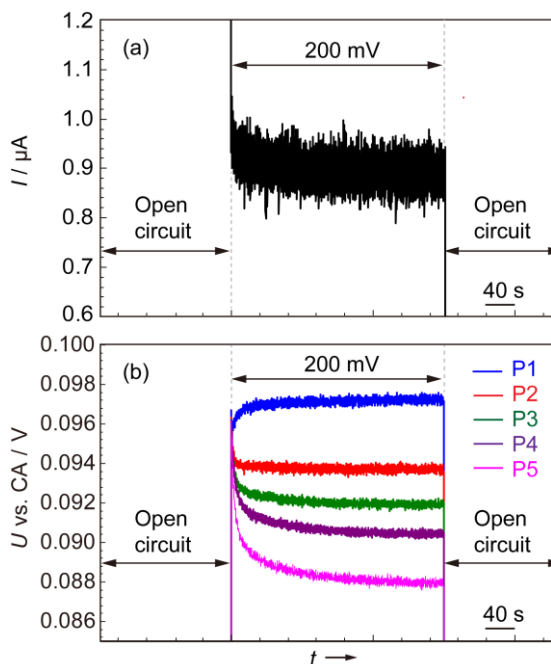The results of Fig. 5 also indicate that the electric current in the redox electrolyte

mainly flows by electron-transfer reactions between R_i and Ox_i or Ox_j, as mentioned earlier. In Fig. 5, the period of time in which the electric current or the potentials of five Pt probes reach the middle between the initial and the stationary values is about 5 s. This is most clearly seen in the potential of a Pt probe named P5. This period of time is much shorter than expected from an assumption that a stationary

Fig. 6. Time courses of the potentials U of five Pt probes vs. CA (cathode) when they were short-circuited and then separated, under the application of 200 mV between anode and cathode.

state is attained by the diffusion of R_i and Ox_i, thus giving support to the above conclusion. In fact, the period of time, t, needed for attaining a stationary state by such diffusion can be estimated from an equation of $t = x^2/D$, where x is the diffusion distance and D the diffusion constant and is calculated to be 40 s when $x = 0.2$ mm and $D = 1 \times 10^{-5}$ cm^2 s^{-1}. In the present work, multi-valent and high-concentration redox couples are used and the diffusion constants for them are expected to be much smaller than 1×10^{-5} cm^2 s^{-1}. In addition, the effective diffusion distance x is also expected, from a cell structure of Fig. 4, to be much larger than 0.2 mm. Therefore, the actual period of time t should be much larger than the above calculated value.

We also investigated the stability of a stationary state. Figure 6 shows how the potentials of five Pt probes changed when these probes were first manually short-circuited for 120 s after the attainment of a stationary state under the application of 200 mV between anode and cathode and then restored to the original state with no short-circuit. The observed potentials of five Pt probes just after the removal of the short-circuit were disturbed by noises but we can see that they each soon (within a few s) returned to the original stationary-state values, confirming that a stationary state is kept stable against fluctuation.

Discussion

Computer simulation about time courses of concentrations of chemical substances in an ORN clearly indicates that an ORN reaches a stationary state after the lapse of a sufficiently long time. Essentially the same results are obtained for ORN's under various reaction conditions and those with different structures from Fig. 1, showing that they give general behavior of ORN's. This result can be explained as follows. Rate equations such as expressed by Eqs. (1) and (2) indicate that reaction and diffusion processes are irreversible and are always forced to approach equilibrium. Thus, such processes in an ORN, which are mutually linked with each other, spontaneously happen and finally reach a state in which they have approached equilibrium to the largest extent as a whole under given conditions and attained a full balance. This is because rates of irreversible processes
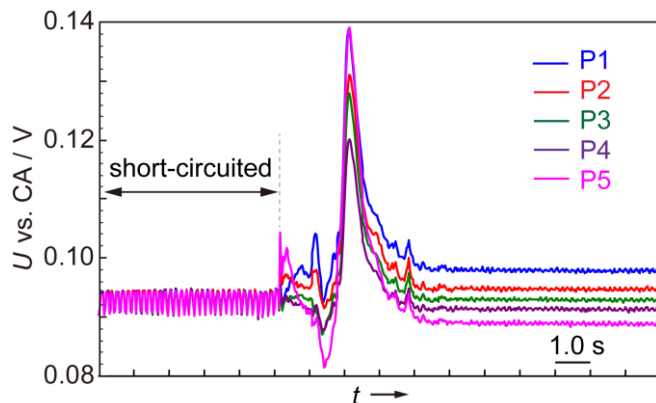
continue to change until they attain a full balance. When irreversible reaction and diffusion processes have attained a full balance, the rates of irreversible processes become constant and the rates of concentration changes for all R_i come to be zero, namely a stationary state is formed.

The above explanation indicates that an ORN necessarily reaches a stationary state and in addition a stationary state thus formed is firmly stabilized by irreversible reaction and diffusion processes and is kept stable against fluctuation. Quantitatively, the time courses of concentrations C_i of chemical substances R_i in an ORN can be described by a set of differential equations,

$$dC_i/dt = f_i(C_1, C_2, \cdots C_i, \cdots C_n), \qquad i = 1, 2, \cdots n \qquad (4)$$

where f_i is a function of C_i and some parameters characterizing an ORN. A stationary state of an ORN is thus given by the following equations

$$dC_i/dt = f_i(C_1^s, C_2^s, \cdots C_i^s, \cdots C_n^s) = 0, \qquad i = 1, 2, \cdots n \qquad (5)$$

where C_i^s is the concentration C_i in a stationary state. According to this formulation, the attainment of a stationary state in an ORN means that Eqs. (4) are converted to Eqs. (5) with time. Equations (4) and (5) for ORN's in the present work are, however, too complex to deal with analytically. The stability of a stationary state can also be investigated by using the Lyapunov function[9] but Eqs. (5) are again too complex. Fortunately, the formation and the stabilization of a stationary state can be explained by considering a function f_{DOB} given by the sum of the absolute values of dC_i/dt in an ORN,

$$f_{\text{DOB}} = \sum_i |dC_i/dt|. \qquad (6)$$

In a stationary state, $f_{\text{DOB}} = 0$ because all irreversible processes have attained a full balance and no concentration change happens, as mentioned above. In a non-stationary state, $f_{\text{DOB}} > 0$. In addition, $|dC_i/dt|$ increases as an ORN goes away from a stationary state, as can be seen from rate equations such as expressed by Eqs. (1) and (2). Thus, f_{DOB} can be illustrated in such a way as Fig. 7 and can be interpreted as representing the degree of balance (DOB) in an ORN. Rate equations such as expressed by Eqs. (1) and (2) also indicate that $|dC_i/dt|$ and hence f_{DOB} spontaneously decrease with time on average. Therefore, Fig. 7 shows that a non-stationary state with $f_{\text{DOB}} > 0$ necessarily returns to a stationary state and a stationary state thus formed is kept stable against fluctuation, as mentioned above.

The time courses of the entropy production rate, σ_1, σ_2 and σ, in Fig. 3 also show that an ORN reaches a stationary state. However, the result of Fig. 3(b) shows that we cannot necessarily say that the σ in an ORN monotonously decreases when it approaches a stationary state, as mentioned earlier. Therefore, we cannot use the σ as a specific quantity to characterize a stationary state of an ORN. The above f_{DOB} can be used as such a quantity, instead of σ. The
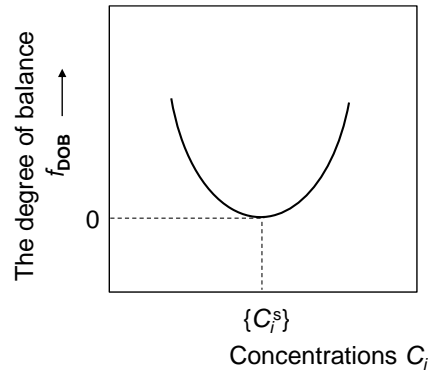

Fig. 7. The degree of balance, f_{DOB}, as a function of the concentrations C_i of chemical substances R_i in an ORN. $\{C_i^s\}$ refers to a set of the concentrations C_i in a stationary state.

above-mentioned properties of f_{DOB} apply, irrespective of whether or not reactions in an ORN are placed under constant thermodynamic forces and whether or not they are in the linear region.

The above argument that an ORN necessarily reaches a stationary state and a stationary state thus formed is kept stable against fluctuation was clearly confirmed by experiments as well, as can be seen from the results shown in Figs. 5 and 6. In particular, the result of Fig. 6 definitely indicates that a destabilized non-stationary state caused by a fluctuation soon returns to the original stationary state.

It has now become clear that a stationary state, which an ORN finally reaches, is firmly stabilized and kept stable against fluctuation. This means that a stationary state of an ORN has ability to organize itself. Such self-organizing ability arises from the fact that irreversible reaction and diffusion processes, which are always forced to approach equilibrium, are fully balanced in a stationary state. Indeed, a stationary state of an ORN is maintained by inflows of source substances at constant rates and a network of chemical reactions but the qualities of it such as self-organizing ability arise from fully-balanced (or continuously-connected) irreversible reaction and diffusion processes. We can see from this argument that irreversible processes and their full balance constitute two necessary conditions for the emergence of self-organizing ability. In relation to this consideration, it is interesting to note that a machine such as a humanoid robot, which can be regarded as a network of processes of transmitting forces or signals driven by external power, has no self-organizing ability, in contrast to an ORN. This is natural if we consider that processes in a machine are of a reversible character, contrary to reaction and diffusion processes, and have no property of spontaneously and irreversibly happening in a particular direction and hence attain no full balance.

A stationary state of an ORN can be easily formed in a prebiotic chemical condition. In addition, a stationary state of an ORN is a non-equilibrium dynamic state and flexible in structure and properties and thus can evolve based on its own self-organizing ability through interaction with the environment. Thus, it has large potential for producing diverse ordered structures in the course of evolution and is expected to have played a fundamental role in the emergence of life on the primitive earth.

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