Dynamics of Interacting Nonlinear Oscillators

Experiments on Chemical Oscillator and Salt-Water Oscillator

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1. Importance of the Tri-phasic Mode

Conversion of chemical energy into vectorial process is quite important in living organisms. Muscle contraction, Flagella motor, active transport in cell membrane; all of these functions are generated with the consumption of chemical energy. According to the theorem of Curie-Prigogine, chemical reaction in isotropic environment can not connect with the vectorial processes. Though this theorem is important, it is much more interesting to know the conditions how chemical reaction can produce vectorial processes with high efficiency. In the present report, we would like to describe that tri-phasic mode is quite stable as a mode of entrainment among three nonlinear oscillators. The presence of tri-phase mode corresponds to the generation of "torque" on a plane. "Torque" produced by the tri-phasic mode can, thus, create vectorial process. We would like to report the presence of the stable tri-phasic mode based on two different experiments, rhythmic chemical reaction and salt-water oscillator, and discuss on the coupling among three nonlinear oscillators based on nonlinear differential equations.

2. Tri-phasic Mode in Interacting Chemical Oscillators

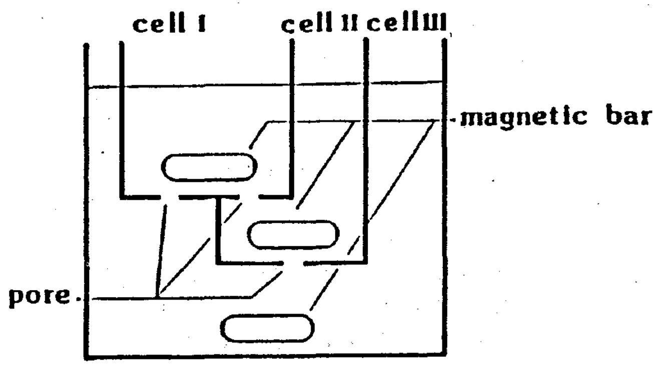

Fig.1 Experimental apparatus for the measurement of interference of three chemical oscillators.
Each pore size: 1mm ϕ.
Reactant: 3M H_2O_2, 0.2M KIO_3,
0.16M $HClO_4$,
0.15M Malonic Acid,
0.015M $Mn(II)SO_4$,
1% starch.
Each magnetic bar rotates in the same speed driven by a magnetic stirrer situated under the apparatus.

The Briggs-Rauscher(BR) reaction[1] is a well-known chemical oscillation as the most visually impressive rhythmic reaction. The BR reaction is the malonic acid by a mixture of hydrogen peroxide and iodate catalyzed by manganous ion in the presence of starch. Under appropriate conditions a stirred batch solution goes through 15 or more cycles of colorless-dark blue. The BR reaction exhibits various nonlinear phenomena such as periodic oscillation, complex oscillation, multi stable states accompanied by hysteresis.[2] We have found that tri-phasic mode is stable when three rhysmic oscillators of the BR reaction interact each other.

Experimental apparatus is show in Fig.1. In this system, we can study the interaction of the oscillators under the condition of the same stirring speed in each reactor.

Fig.2 shows the result of the experiment, suggesting each oscillation are entrained in a tri-phasic mode after the 2 or 3 cycles of the oscillations. We have found that this mode is quite stable against perturbations, such as by shaking the experimental apparatus.

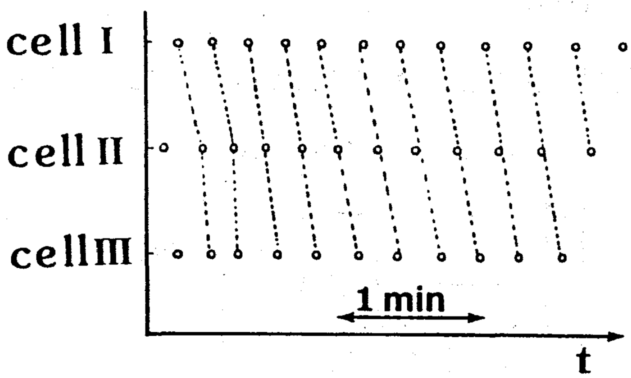

Fig.2 Tri-phasic mode of entrainment among three chemical oscillators. The open circle indicates the appearance of dark bulk color in the reactanting solution.

3. Tri-phasic Mode in Interacting Salt-Water Oscillators

When a plastic cup of salt-water with a pore-diameter less than 2 mm on the bottom-wall is partially submerged in a beaker of pure-water, rhythmic change of the water flow is generated and continues for several hundred cycles.[3]

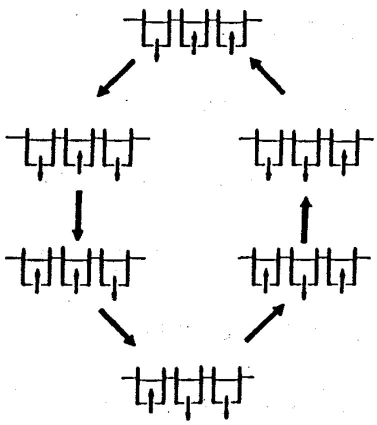

Fig.3 Schematic representation of the manner of entrainment among three equivalent salt-water oscillators dipped in a single outer vessel.

Interference between the salt-water oscillators is interesting, because various types of spatio-temporal pattern are induced when the oscillators interact each other.[4] In Fig.3 the mode of interference among the three salt-water oscillators is schematically given. This mode is generated when three equivalent cups with the same pore-size on their bottom are immersed into a vessel of pure-water. Here, the level of pure-water in the outer vessel is the connecting factor among the oscillators. The tri-phasic mode is stable because the level of water in the outer vessel remains essentially constant.

Based on the Navier-Stokes equation, the experimental mode of interference or entrainment can be reproduced by numerical simulations with appropriate approximations.[3] Eq.(1) gives the equation to interpret the interference among salt-water oscillators, when cups with the same size, but not necessarily with the same pore-size, are immersed into a single outer vessel.

$$\ddot{X}_j + A_j|\dot{X}_j|\dot{X}_j - B_j\dot{X}_j + C_jX_j + D_j\sum_{k\neq j}^{\text{all cups}} X_k = \text{const.} \quad (1)$$

Where X_j is the height of the level of salt-water in the j_{th} cup. The second term represents the degree of the pressure-loss induced when the water-flow goes though the pore of the cup and the third term refers to the acceleration of the flow due to the buoyancy force which is partly diminished by the viscosity of the fluid. The fourth term shows the effect of the force of gravity, caused by the change in the level of water of the j_{th} cup. The last term in the left of the equation the corresponds to the pressure of gravity arising from the change in the level of water in the outer vessel due to the water flow in all the cups except the j_{th} one. The parameter D_j is a measure of the relative ratio between the surface are of a cup and the free surface area of the outer vessel. Fig.4 shows the result of the simulation of the entrainment among three salt-water oscillators, cup-I,II,III. For simplicity, only the changes of the direction of the flow are given. At the first stage of the oscillations, the frequency of the each oscillator fluctuates greatly. After ca. 2 minutes, the mode of the oscillation is locked with the phase difference of $2\pi/3$ each other. The actual experiments of the salt-water oscillators afford essentially the same behavior as in the numerical calculation.

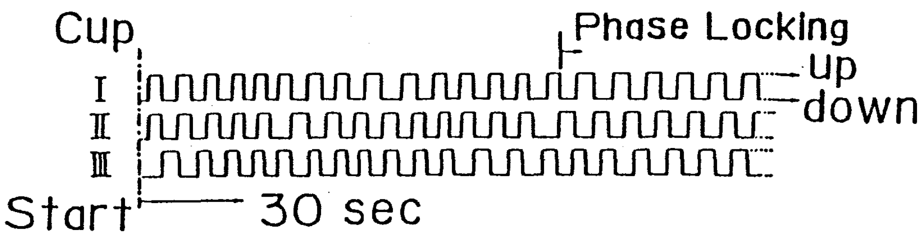

Fig.4 Computer simulation for the entrainment among three salt-water oscillators.
The pore diameters of the cups are 1.000,1.004, and 0.997mm

4. On the Stability of the Tri-phasic Mode

When we replace the second term in eq.(1) by $\dot{X}^3$ as an odd function of $\dot{X}$, the following equations are derived as a model to describe the interacting three equivalent oscillators.

$$\ddot{X} - (a-b\dot{X}^2)\dot{X} + cX + d(Y+Z) = 0$$

$$\ddot{Y} - (a-b\dot{Y}^2)\dot{Y} + cY + d(Z+X) = 0 \qquad (2)$$

$$\ddot{Z} - (a-b\dot{Z}^2)\dot{Z} + cZ + d(X+Y) = 0$$

In the case of the salt-water oscillator, the parameter d is proportional to the relative ratio between the surface area of a single cup and the surface area of the outer vessel. We define W as follows.

$$W = X + Y + Z \qquad (3)$$

From eqs.(2) and (3),

$$\ddot{W} - a\dot{W} + b\dot{W}(\dot{X}^2+\dot{Y}^2+\dot{Z}^2-\dot{X}\dot{Y}-\dot{Y}\dot{Z}-\dot{Z}\dot{X}) + 3b\dot{X}\dot{Y}\dot{Z} + (c+2d)W = 0 \qquad (4)$$

When W is zero(W=0). All of the terms except $\dot{X}\dot{Y}\dot{Z}$ vanish. If we assume that X,Y and Z changes in sinusoidal manner, X,Y and Z are expressed as follows.

$$X = \sin(\omega t)$$

$$Y = \sin(\omega t + \theta_1) \qquad (5)$$

$$Z = \sin(\omega t + \theta_2)$$

When W=0, θ_1, θ_2 have the following relationship.

$$|\theta_1 - \theta_2| = \frac{2}{3}\pi \text{ or } \frac{4}{3}\pi \tag{6}$$

This indicates that a solution with tri-phasic mode is present in eq.(4), though detailed analysis of stability of the solution is necessary.

Similar discussion stands for the tri-phasic entrainment in the chemical oscillator. It has been well established[5] that the essential feature of spatio-temporal structure in the oscillatory chemical reaction such as the Belousov-Zhabotinskii reaction, can be described using the Bonhoeffer-van der Pol equation.

$$\begin{aligned} \tau u_t &= \varepsilon^2 \nabla^2 u + u - \frac{u^3}{3} + v \\ v_t &= \nabla^2 v - \gamma u \end{aligned} \tag{7}$$

We convert the variables (u,v) to (x,y).

$$\begin{aligned} u &\rightarrow x \\ v &\rightarrow -y \end{aligned} \tag{8}$$

If $\nabla^2 v \equiv \nabla^2(-y)$ is negligible small, eq.(9) is obtained. This corresponds to the assumption that only the diffusion of activation u(or x) is important.

$$\begin{aligned} \tau x_t &= \varepsilon^2 \nabla^2 x + x - \frac{x^3}{3} - y \\ y_t &= \gamma x \end{aligned} \tag{9}$$

Let us consider the experiment as in Fig.3 and 4. The diffusion of x is generated though the pore, and the concentrations of x and y are homogeneous in each reactor owe to the continuous mixing. If we define x_i and y_i as the concentrations of the reactant in the i_{th} reactor, and if we consider the simplified case as $\tau = \varepsilon = \gamma = 1$, the following equations are derived.

$$\begin{aligned} \frac{dx_i}{dt} &= x_i - \frac{1}{3}x_i^3 - y_i + C(\sum_{j \neq i} x_j - 2x_i) \\ \frac{dy_i}{dt} &= x_i \end{aligned} \tag{10}$$

$$i, j = 1, 2, 3$$

Where C is a parameter corresponding to the area of the pore between the reactors. Eq.(10) is reformed as follows.

$$\frac{d^2y_i}{dt^2} - \frac{dy_i}{dt} + \frac{1}{3}\left(\frac{dy_i}{dt}\right)^3 + y_i + C\left(\sum_{j \neq i}\frac{dy_j}{dt} - 2\frac{dy_i}{dt}\right) = 0 \tag{11}$$

$i,j = 1,2,3$

From the above equations (three differential equations), we obtain.

$$\frac{d^2(y_1 + y_2 + y_3)}{dt^2} + \frac{d(y_1 + y_2 + y_3)}{dt}$$

$$+ \frac{1}{3}\left\{\left(\frac{dy_1}{dt}\right)^3 + \left(\frac{dy_2}{dt}\right)^3 + \left(\frac{dy_3}{dt}\right)^3\right\} + y_1 + y_2 + y_3 = 0 \tag{12}$$

Here, one can see that similar discussion stands for in this case as that in the tri-phasic entrainment of the salt-water oscillators. It is clear that tri-phasic mode is a general solution for the interacting system of three chemical oscillators. Further study concerning the mathematical stability of the tri-phasic mode is awaited.

At the last, we would like to mention on the direction of "torque" of the tri-phasic made, or the moving direction of the "phase-wave". As for the tri-phasic mode of the entrainment of three oscillators (A,B and C), there are two degenerate states of clockwise and anticlockwise modes, i.e., bistable modes, (see Fig.5).

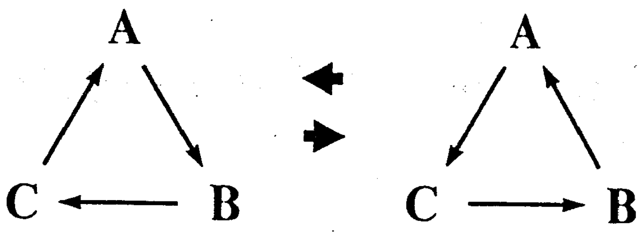

Fig.5 Bistability of the mode of tri-phasic entrainment

As is expected from the above discussion, the mode is critically dependent on the initial condition. This system is regarded as a new kind of element of 'memory'.

Acknowledgment

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