

Spatial Symmetry Breaking in the Revival Wave of the Belousov-Zhabotinsky Reaction Containing 1,4-Cyclohexanedione

C. Basavaraja, Na Ri Kim, Hyun Tae Park, and Do Sung Huh*

Department of Chemistry and Institute of Functional Material, Inje University, Kimhae, Kyungnam 621-749, Korea

*E-mail: chemhds@inje.ac.kr

Received February 11, 2009, Accepted March 5, 2009

Complex breakup behavior in the revival wave has been observed in the Belousov-Zhabotinsky (BZ) reaction system containing 1,4-cyclohexanedione (1,4-CHD) in the dish divided into two compartments with a sliding window. A same reaction mixture is poured into the two compartments individually with time difference. Wave propagation exhibited different behavior in the revival wave of the reaction system. This was largely dependent on the progress time prior to the pouring into each compartment and on the gap between the times of pouring into the two compartments. The revival wave in the reaction system is induced spontaneously as a new wave train with a long time lag after the disappearance of the initially induced wave. A thoroughgoing study of the chaotic breakup of propagating chemical wave train was to be possible since the revival wave has a longer wavelength, clearer wave-train patterns, and longer duration period.

Key Words: BZ-Reaction, Chaotic breakup, Divided-dish, Reaction-diffusion, Revival wave

Introduction

Chemical wave formation is one of the central problems in the study of modern macroscopic reaction kinetics. In the last decade, pattern formation in perturbed reaction-diffusion media has attracted increasing attention from scientists working in several disciplines. Specifically, there have been numerous studies on the problems which may influence to the *reaction-diffusion* phenomena,^{1,2} such as chemically reacting fluids,^{3,4} interacting populations and ecosystems,⁵ and systems with propagating fronts.^{6,7} By definition, there are no flows involved in a reaction-diffusion system; any mixing between the different species is achieved solely through diffusion. The flow of fluids significantly enhances the mixing of reactive substances mostly in the real systems. Occurrences like this are beyond diffusion alone. The advective mixing of a reactive substance itself has a significant influence on pattern formation process. This is particularly important when we consider the recent studies showing that a mixing can be chaotic even for simple, well-ordered laminar fluid flows.^{8,9} In addition, there have been several theoretical and numerical studies considering the influence of chaotic advection on chemical and biological pattern-forming systems.¹⁰⁻¹³ And studies have also indicated that chaotic mixing in reacting systems typically produces filamentary patterns with the filaments mimicking the structures used to describe chaotic mixing in the flows.¹⁴⁻¹⁶

The BZ reaction is one of the systems which have laid a solid foundation in understanding many natural phenomena through the reaction-diffusion (RD) mechanism. One such phenomenon is the periodic pattern formation among living systems.¹⁷ Its importance is the main reason behind the extensive studies which delved on this class of chemical system. An important example is the system's wide variety of biological implications. Zhaikin and Zhabotinskii¹⁸ were the first to describe propagating waves in the BZ reaction system.

Such a system shows periodic or complex spatio-temporal dynamics, including chaotic behavior.¹⁸⁻²² For more than three decades, the BZ reaction has been used to study various non-linear phenomena such as oscillation, excitability, chaos, pattern formation, and wave propagation. These phenomena are found within the realm of biology over a broad range of complexity. The mechanism of pattern formation is one of the key issues in nonlinear chemistry and physics, developmental biology, and ecology. In terms of experimentally induced spatial phenomena, the used substances are known and the products are detectable. In this point, it is relatively easy to study the molecular mechanism behind the pattern formation. For this reason, it has become a model system used for studying many physical, chemical, and biological pattern phenomena obtained under conditions far from equilibrium. Since the patterns are created by the internal dynamics of the system, the process is called spatiotemporal self-organization. This is common in most of the natural systems.^{18,23-30}

The BZ reaction means the oxidation reaction of an organic substrate through the use of bromate under the action of a catalyst (a metal-ion redox couple) in an acidic environment.^{31,32} In the widely studied or traditional version of the BZ reaction, malonic acid (MA) is used as the organic substrate. However, the reaction system has an inevitable drawback producing carbon dioxide gas from malonic acid, which induces hydrodynamic disturbances in the long term observations of the spatial pattern generations and propagating waves. For this reason, a variety of new substrates have been introduced to replace MA in the aim of overcoming the problem. Although many substrate analogues of the traditional BZ system are known, only a few has been utilized so far relating with the studies of wave propagation.³³ The gas-free BZ-system using diketonic compound such as 1,4-cyclohexanedione (1,4-CHD) instead of malonic acid has attracted increasing attention in the last decade. Another attractive property of the CHD system is complicated non-linear pheno-

mena that account for wave stacking and merging^{34,35} not observed in the traditional BZ system and its complicated response to the illumination of visible light. It means that an intensive investigation on wave propagation in the BZ system containing 1,4-CHD can reveal new dynamic phenomena previously undetected in the traditional BZ system.

Recently, Huh *et al.* have discovered an unusual wave train in the bromate-1,4-cyclohexanedione-ferroin-sulfuric acid reaction system which induces two types of wave with a long time lag.³⁶⁻³⁹ A new wave is induced as a secondary or revival wave after the complete disappearance of an initially induced wave in the form of a concentric target pattern differently from the initially induced wave showing a spiral pattern with a high wave frequency. The variation of the reactant concentrations significantly altered the initiation and duration of the initial waves in the system. However, the variation has a small effect on both the dormant times and the appearance of the secondary waves. The dormant times pertain to the transient period prior to the appearance of secondary waves. The revival wave pattern achieved through a chemical effect and an environmental stimulus, such as the illumination of visible light,^{37,38} has likewise been reported. Previous investigations have suggested that the reaction mechanism governing revival wave formation in the system might be different from the initial waves. In addition, the initiation of the two waves might be induced through different reaction mechanisms in the same initial reaction system.³⁶⁻³⁹

In a series of reports by Steinbock *et al.*,⁴⁰⁻⁴³ the anomalous dispersion in excitable media of the wave dynamics in a thin capillary tube filled with CHD-BZ solutions has been presented. The study demonstrated the existence of an instability in which an initially periodic wave train decays into an aperiodic one ("pulse bunching"). In doing so, evidence for the interaction between successive pulses have been provided. These pulses either induce stable bound wave packets formation or facilitate apparent merging of pulses in back-to-front collisions.⁴⁰ Furthermore, three-dimensional scroll waves that can rotate around filaments were observed. This finding is of prime importance because it broadens current understanding of the topological constraints governing basic characteristics of scroll wave filaments.⁴¹

In this study we have examined a chaotic breakup of the propagating wave-train by using a half-divided Petri dish partitioned into two compartments with a sliding window. The reaction system of bromate-1,4-cyclohexanedione-ferroin-sulfuric acid which induces two wave patterns of the initial and the revival wave has been employed. Two stages of wave pattern are induced in each compartment by pouring two suspensions of same concentration but with time gap pouring into each compartment. The chaotic breakup of propagating wave-train was induced spontaneously in the stage of the revival wave by opening the sliding window, and the breakup of the wavelength of the revival wave depended on the concentration of $[Bromate]_0$ and $[1,4-CHD]_0$, and the time gap of pouring between the two compartments.

Experimental Section

The propagating chemical wave was observed in a thin

layer of cation exchange resin loaded with the metal catalyst of ferroin. The analytical grade cation-exchange resin (Dowex 50W-X4) of mesh 400 (bead diameter 40 - 70 μm) was purchased from the Sigma company. Because the purchased cation resin is slightly acidic, these beads were first washed several times by distilled water to neutralize to load ferroin catalyst to the cation beads. Afterwards, the resin was loaded with ferroin by mixing a measured quantity of resin beads with a specific volume of ferroin solution and by gently stirring for an hour.³⁸ The amount of ferroin immobilized by the cation resin is then calculated from the difference in the ferroin concentration of the bulk solution. Stock solutions were prepared from NaBrO_3 (Aldrich, 99%), 0.6 M, dissolved in distilled water; 1,4-cyclohexanedione (Fluka, 98%), 0.2 M, dissolved in distilled water; sulfuric acid (Aldrich, 98%), 2 M, dissolved in distilled water. 0.025 M $\text{Fe}(\text{phen})_3\text{SO}_4$ was purchased from Sigma (>99%). All the reagents were used in their commercial grade without further purification.

To begin our experiments, two reaction mixtures with same concentration of reagents and same amount of resin beads loaded with same concentration of ferroin were prepared with long time gaps for the two separate compartments of the half-divided Petri dish (10 cm in total diameter) as 25.0 mL reaction mixture containing 2.5 g of beads for each compartment. The concentration of ferroin is calculated by using the total amount of ferroin contained in the 2.5 g cation resin divided by the volume of the reaction mixture of 25 mL. The reaction mixtures were poured into each compartment of the half-divided dish with time gap of pouring, which formed a uniform thin film of resin on the bottom of the half-divided Petri dish. The thickness of the bead film was calculated from the volume of the resin divided by the area of the bottom of the Petri-dish, which gave us a magnitude of 1 ± 0.3 mm. The half-dish was manufactured by special order from the Yuil Scientific Co. It was designed to prevent mixing the solution from one compartment to the other before opening the window. The partition has a thin sliding window 5 mm in length. All experiments were monitored with a CCD camera

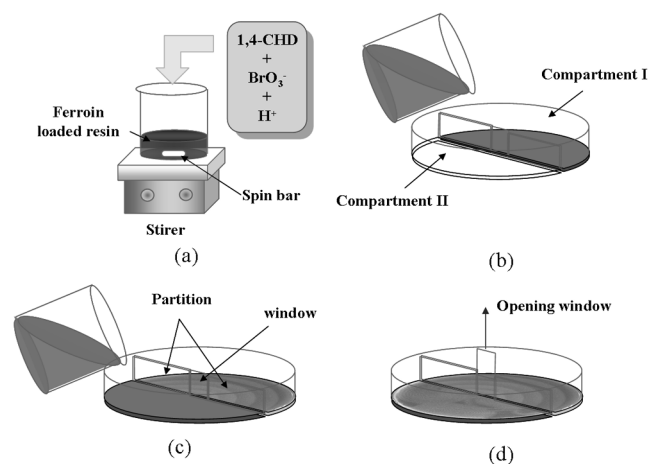


Figure 1. The overall scheme of the experimental process in a half-divided Petri dish system. (a) Mixing of reactants with ferroin loaded resin, (b) the mixture solution is poured at the compartment I at first, (c) another mixture solution with same concentration is poured at the compartment II, and (d) opening the window.

(Sony, SSC-370) equipped with a zoom lens (Niko, 1.4X). The CCD camera was connected to a personal computer running a frame grabber (Flash Point, Opimus 6.1). The wave features, including the wave velocity and the wavelength were measured by time processing in the computer. The schematic procedure of experimental system is presented in Figure 1.

To generate two wave patterns within the reaction system, the two identical suspensions of the reaction solution were poured into the compartments at different times. When a revival wave was induced in the first compartment (compartment-I), the second reaction mixture was added to the second compartment (compartment-II). The sliding window was carefully opened once the revival wave was observed in compartment-I to allow the interchange of the two mixtures in the system by liquid diffusion. The state of wave behavior in the dish may be expressed as $t + \Delta t$, where t is the processing time of the mixture in compartment-I and Δt is the time interval between the additions to the two compartments; consequently, $t + \Delta t$ corresponds to the processing time of the reaction mixture in compartment-II. A reaction time of 1500 min induces a revival wave completely in compartment-I and 60 min induces an initial wave in compartment-II, which corresponds to $t = 1500$ min and $\Delta t = -1440$ min.

Results and Discussion

Revival wave induced by the system in a non-divided dish.

Before the experiments in the half-divided dish, a test experiment was performed in a non-divided dish to identify a revival wave in the system. After an induction period of about 60 min, reddish spots began to appear spontaneously over the surface of the resin layer. Spiral waves then initiated from each spot, as shown in Figure 2a. At this stage (stage I), these waves show an irregular spiral pattern with a high frequency, similar to the results obtained in the BZ reaction using malonic acid.³⁹ A characteristic behavior of the wave pattern was obtained in the system as the reaction time was extended. The initially obtained wave disappeared after about 90 min, as shown in Figure 2b. This stage (stage II) persists for about 1000 min, without the generation of any wave pattern. A new wave is subsequently spontaneously induced, as shown in Figure 2c. The new waves obtained during this stage (stage III) are the revival waves in the system, and are markedly different from those observed initially and shown in Figure 2a. There occurs a concentric target pattern with a regular propagating front, and the wavelength is long relative to that of the initially induced wave. The typical characteristics of the two waves obtained in the system are summarized in Table 1. The detailed behavior of the reaction system in a non-divided Petri dish has been discussed in detail by Huh *et al.*³⁵

Propagating revival wave in a divided dish. Figure 3 presents a series of snapshots showing the breakup of revival waves in the bromate-1,4-cyclohexanedione-ferroin system in the half-divided dish with $\Delta t = -1440$ min after the two compositions are poured into the dish separately with a 60 min interval. Figure 3a shows the state before the sliding window is opened, while Figure 3b, c, and d present the state after

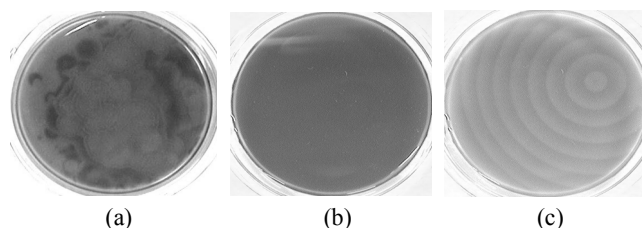


Figure 2. Revival wave patterns obtained in the 1,4-CHD/ BrO_3^- /ferroin/ H^+ system. The initial condition corresponds to $[\text{1,4-CHD}]_0 = 0.05$ M, $[\text{BrO}_3^-]_0 = 0.15$ M, $[\text{ferroin}]_0 = 1.5 \times 10^{-3}$ M, and $[\text{H}^+]_0 = 1.0$ M. The time of process is 80 min in (a), 300 min in (b), and 1300 min in (c) after pouring into the Petri dish.

Table 1. Typical characteristics of propagating wave behavior obtained in the 1,4-CHD/ BrO_3^- /ferroin/ H^+ reaction system

	Initial wave	Break period	Revival wave
Wave pattern	mixed spiral	no wave	concentric target
Induction time (min)	60 - 80	180-230 ^{a)}	800-1200 ^{b)}
Wavelength (mm)	1.5-2.5	no wave	10-20
Duration (min)	150-250	800-1200	1200-2400

^{a)}Sum of the induction period and duration of the initial wave

^{b)}Corresponding the break period

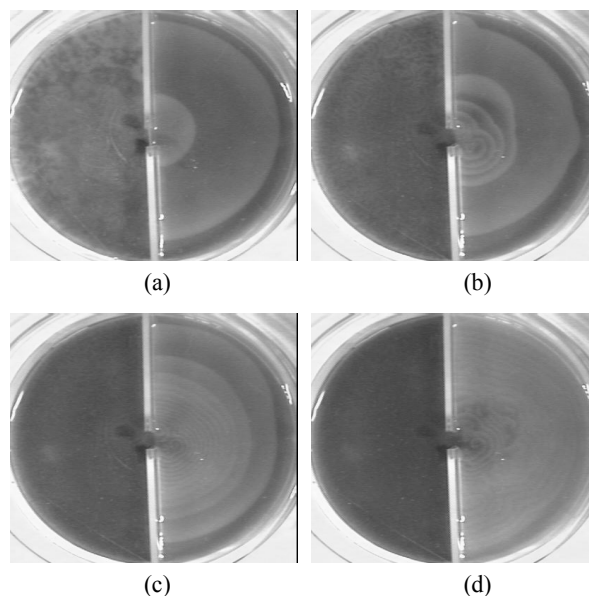


Figure 3. Propagating waves obtained at different times in a divided BZ system when the concentrations on both sides of the dish are $[\text{1,4-CHD}]_0 = 0.05$ M, $[\text{BrO}_3^-]_0 = 0.13$ M, $[\text{ferroin}]_0 = 1.5 \times 10^{-3}$ M, and $[\text{H}^+]_0 = 1.0$ M with $\Delta t = -1440$ min. The time of process in compartment I is (a) $t = 1500$ min, (b) $t = 1530$ min, (c) $t = 1545$ min, (d) $t = 1575$ min.

opening. The waves obtained in compartment I are revival waves and those found in compartment II are the initial waves.³⁸ Figure 3b and c illustrate the state of the half-divided dish at $t = 1530$ and 1545 min, respectively. The changes obtained in both compartments are hardly noticeable and disappeared gradually as shown in Figure 3c. Figure 3d displays a clear breakup of the wave-train into various sizes at $t = 1575$ min in compartment I. It is therefore assumed that the

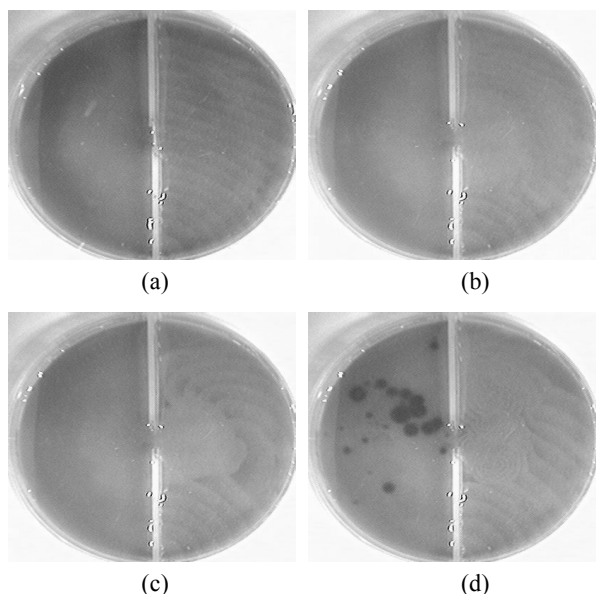


Figure 4. Distinctive breakup in the revival wave obtained by the variation of the reaction time of both the components in a divided BZ system when the concentrations on both sides of the dish are $[1,4\text{-CHD}]_0 = 0.05\text{ M}$, $[\text{BrO}_3^-]_0 = 0.13\text{ M}$, $[\text{ferroin}]_0 = 1.5 \times 10^{-3}\text{ M}$, and $[\text{H}^+]_0 = 1.0\text{ M}$ with $\Delta t = -1020\text{ min}$. The reaction time of process in compartment I is (a) $t = 1070\text{ min}$, (b) $t = 1094\text{ min}$, (c) $t = 1110\text{ min}$, and (d) $t = 1145\text{ min}$.

pattern changes can be obtained in both compartments. It has been established that CHD-BZ is quite different from the classical BZ system in terms of chemical kinetics and nonlinear dynamics. Significantly, the dispersion relation for the CHD-BZ system wave trains is quite unusual as reported by Steinbock *et al.*⁴² The CHD-BZ spatio-temporal behavior is associated with the stable focal characteristics of the steady state. This is considered unusual compared to the nodal steady state in the classical BZ system. Therefore, relaxation behavior is not merely exponential but a periodic function of time. The reaction intermediating among different reaction kinetics could interchange by diffusing through the window. In addition, this can produce a dynamic wave formation instability that creates a functional heterogeneity of the medium in compartment I. This might have induced the breakup of revival waves in compartment I, as shown in Figure 3d. Therefore, an unusual system behavior with reaction kinetics differing with reaction progress seems to be a major factor in inducing an easy breakup of a propagating wave-train.

Another type of breakup in revival waves is also achieved by varying the reaction time in each of the two compartments. Figure 4 shows the wave propagation with $\Delta t = -1020\text{ min}$. As illustrated in Figure 4b, an initial wave was not induced before opening the sliding window in compartment II, though a kind of revival wave was already induced in compartment I. Due to the short reaction time, it can be observed that the wavelengths of the revival wave in compartment I are also shorter. Figure 4c shows a continuous no-wave state in compartment II. Moreover, some wave-breaks in the revival wave appearing in compartment I only have minute changes in wavelength. Figure 4d proved that an initial wave has appeared in compartment II and the breakup of the revival wave extended

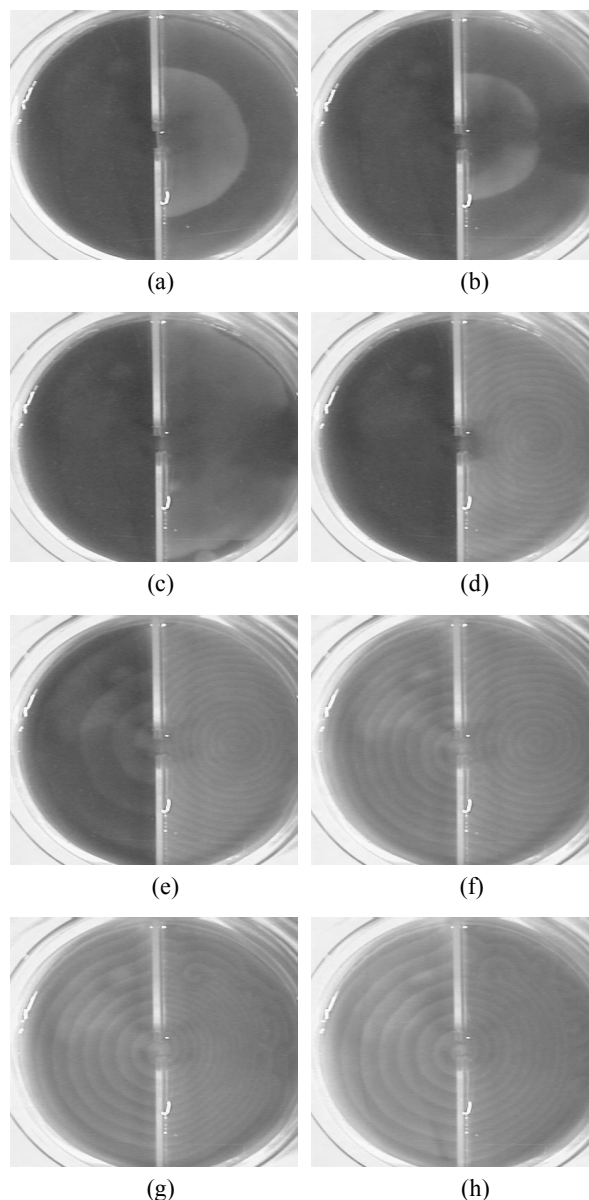


Figure 5. Pattern of wave propagation obtained in a divided BZ system. The initial condition is $[1,4\text{-CHD}]_0 = 0.05\text{ M}$, $[\text{BrO}_3^-]_0 = 0.13\text{ M}$, $[\text{ferroin}]_0 = 1.5 \times 10^{-3}\text{ M}$, and $[\text{H}^+]_0 = 1.0\text{ M}$ with $\Delta t = -500\text{ min}$. The reaction time of process in compartment I is (a) $t = 1350\text{ min}$, (b) $t = 1415\text{ min}$, (c) $t = 1430\text{ min}$, (d) $t = 1445\text{ min}$, (e) $t = 1505\text{ min}$, (f) $t = 1550\text{ min}$, (g) $t = 1605\text{ min}$, and (h) $t = 1640\text{ min}$.

gradually into compartment I. The induction of an initial type wave in compartment II is not much affected by the reaction mixture that induces a revival wave.

However, these time gaps in pouring did not always induce wave-breaks in the propagating wave-train of the revival wave. Figure 5a illustrates a state in compartment II in which the initially induced wave disappeared completely. The wave pattern obtained in compartment I is exactly the same as shown in Figure 4. Huh *et al.* indicate that the break-period of the 1,4-CHD system does not induce any type of wave. Furthermore, it sustains long periods at a reduced metal catalyst state with red color.³⁸ In other words, the medium during break-period is unexcitable. Figure 5b, c, and 5d exhibit the

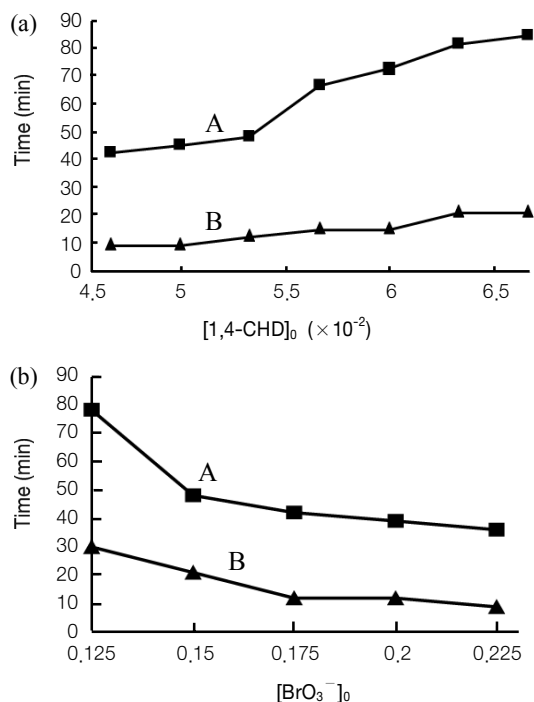


Figure 6. Time lapsed in the appearance (line A) and the disappearance (line B) for break up wave at different concentrations of $[1,4\text{-CHD}]_0$ in (a) and $[\text{BrO}_3^-]_0$ in (b), respectively. The other concentrations are the same as those indicated in Fig. 3.

states after opening the window at 65, 80, and 95 min episodes, respectively. Regardless of the time episodes, the break-period continues in compartment II and the wavelengths of the propagating revival wave increased rather differently. The results shown in Figure 4 imply an impossibility of wave-breaks in this system. However, a major change in wave patterns has occurred in Figure 5e. The revival wave in compartment I changed into a very dense wave by accommodating itself through a new wave induction in compartment II. This new wave can be interpreted as a kind of revival wave since its reaction time and break-period in compartment II matches the general break-period in a non-divided dish. Afterwards, the propagation of the revival wave in compartment I adopted a spiral wave behavior as shown in Figure 5f, g, and h. Lastly, another kind of spiral wave which broke into a chaotic pattern was obtained as shown in Figure 5h.

The results shown in Figure 5d-f emphasize the importance of compartment II's excitability in inducing the spiral breakup of the revival wave in compartment I. This can be interpreted by taking into account the reaction mechanism for unusual wave behavior of the 1, 4-CHD reaction system suggested by Huh *et al.*³⁷ The dynamic instability for the breakup in compartment I cannot be interpreted by using the time gap in pouring the mixture between the two compartments alone. Moreover, there is no consistency observed in the results of Figure 3, 4, and 5 since time difference of pouring between the two compartments is 1440 min in Figure 3, 1020 min in Figure 4, and 500 min in Figure 5, respectively. Moreover, a long time gap in pouring between the two compartments could bring about a large difference of chemical kinetics in the two mixtures. Although wave-breaks were not observed at

first stage, a spontaneous spiral breakup in compartment I was obtained in Figure 5h, which occurred a long time after opening the window.

Effect of $[\text{BrO}_3^-]_0$ and $[1,4\text{-CHD}]_0$ on the revival wave in a half-divided dish. The concentration dependence of $[\text{BrO}_3^-]_0$ and $[1,4\text{-CHD}]_0$ were closely examined to determine the appearance and disappearance of a revival wave by varying the reaction condition ranging from 0.4 to 0.9 and 0.04 to 0.09 M, respectively, and beyond these there were no revival waves observed in the system. The revival wave's time period for appearance (line A) and disappearance (line B) are shown in Figure 6, whereas Figure 6a and b corresponds to $[1,4\text{-CHD}]_0$ and $[\text{BrO}_3^-]_0$ respectively. Figure 6a illustrates that, as $[1,4\text{-CHD}]_0$ increases, line B increases gradually while line A remains more or less the same. The values of line B at a lower concentration was initially 40 min which later became 70 min and even further to 80 min at a higher $[1,4\text{-CHD}]_0$. The magnitude between line A and line B increases in proportion to $[1,4\text{-CHD}]_0$ boost. On the contrary, the values of line B at a lower $[\text{BrO}_3^-]_0$ is higher compared to line A. Similarly, line A decreases as $[\text{BrO}_3^-]_0$ is increased as shown in Figure 6b. The magnitudes between line A and line B appear to be inversely proportional to a higher $[\text{BrO}_3^-]_0$. In general, the magnitudes remain more or less the same but decrease at much higher $[\text{BrO}_3^-]_0$. In sum, the magnitudes, when compared to both $[1,4\text{-CHD}]_0$ and $[\text{BrO}_3^-]_0$, are in contrast with each other. In other words, the amplitude of these two lines increases as $[1,4\text{-CHD}]_0$ boosts and decreases the rise in $[\text{BrO}_3^-]_0$. The findings conclude that the amplitude of these two lines increases at a low $[\text{BrO}_3^-]_0$ and high $[1,4\text{-CHD}]_0$.

Earlier, Huh *et al.*³⁷⁻³⁹ have explained the unusual wave behavior in a 1,4-CHD reaction system by dividing two reaction mechanisms. The induction of the revival wave is explained by the required reaction time in producing an available concentration of H_2Q from 1,4-CHD and Br-CHD .^{44,45} Simply stated, it means that the reaction mixture of the 1,4-CHD reaction system is unexcitable in inducing a wave in the break-period. Though the condition above is suited to a particular environment, the study of chaotic breakup remains a special and complex case unique from other systems reported. It is presumed that the unexcitable reaction mixture has a greater influence on the break up wave. This reaction is regardless of the difference between the reaction kinetics of the two compartments and may also be attributed to the difference of pouring time. Opening the window allows the merging of reaction mixtures with different dynamics. Hence, this scenario implies that hydrodynamic disturbances couples with the dominant reaction-diffusion dynamics of the system. In contrast to the intensively studied CHD-UBO, the latter is more complex due of its dynamic behavior which involves additional reaction such as the ones between CHD, H_2Q , HBrO_2 , and ferroin.

Conclusion

The chaotic breakup of the propagating wave-train in a Petri dish partitioned with two compartments using a modi-

fied BZ reaction system was examined. This system induces two types of patterns, the initial and the revival wave. In lieu of the results generated, further studies are necessary to clarify the kinetics of the CHD-BZ reaction. Research on this area will also lead to the formulation of an appropriate reaction model capable of reproducing the observed wave phenomena. On the other hand, the results of this study serve some significance in light of previous research that has established that mixing can be chaotic even for simple and well-ordered laminar fluid flows. However, in most real systems, fluids flow significantly enhances the mixing of reactive substances beyond diffusion alone. This advective mixing has a significant influence on the pattern formation process. Recent qualitative and quantitative studies have considered the influence of chaotic advection on chemical and biological pattern-forming systems. In particular, studies have shown that chaotic mixing in reacting systems usually produces filamentary patterns. The filaments mimic the structures used to describe chaotic mixing in the fluid flows. Hence, this indicates that the breakup is caused by the instabilities of the excitable medium in the stage of revival wave stage. This is induced by an inhomogeneity of the reaction system by diffusion of the reaction mixture for the initial wave. Nevertheless, there has been a great deal of interest in understanding how patterns form in nonlinear dynamical systems. To illustrate, there have been numerous studies on reaction-diffusion, including chemically reacting fluids, interacting populations and ecosystems, and systems with propagating fronts. A detailed study seeking to understand the experimental process that reinforces recent theoretical and numerical studies is in progress. Studies have indicated that the tools used to describe chaotic mixing can be applied to analyze and predict patterns in a reacting flow, such as opening window as in the case in this experiment.

Acknowledgments. This work was supported by the Korea Research Foundation Grant funded by the Korean Government (MOEHRD) (KRF-00042007070-00).

References

- Winfrey, A. T. *Science* **1972**, *175*, 634.
- Showalter, K. J. *Chem. Phys.* **1980**, *73*, 3735.
- Cantrell, R. S. *SIAM Rev.* **1996**, *38*, 256.
- Handbook of Crystal Growth*; Hurler, D. T. J., Ed.; North-Holland: Amsterdam, 1993; Vol. 1B.
- The Theory and Applications of Reaction-diffusion Equations: Patterns and Waves*; Grindrod, P., Ed.; Clarendon Press: Oxford, 1996.
- Diffusion and Reactions in Fractals and Disordered Systems*; Ben-Avraham, D.; Havlin, S., Eds.; Cambridge University Press: Cambridge, 2000.
- Neufeld, Z.; Kiss, I. Z.; Zhou, C. S.; Kurths, J. *Phys. Rev. Lett.* **2003**, *91*, 084101.
- Scheuring, I.; Karolyi, G.; Pentek, T. T. A.; Toroczka, Z. *Freshwater Biol.* **2000**, *45*, 123.
- Abel, M.; Cencini, M.; Vergni, D.; Vulpiani, A. *Phys. Rev. E* **2001**, *64*, 046307.
- Abel, M.; Cencini, M.; Vergni, D.; Vulpiani, A. *Chaos* **2002**, *12*, 481.
- Cencini, M.; Torcini, A.; Vergni, D.; Vulpiani, A. *Phys. Fluids* **2003**, *15*, 679.
- Tel, T.; de Moura, A.; Grebogi, C.; Karolyi, G. *Phys. Rep.* **2005**, *413*, 91.
- Ronney, P. D.; Haslam, B. D.; Rhys, N. O. *Phys. Rev. Lett.* **1995**, *74*, 3804.
- Leconte, M.; Martin, J.; Rakotomalala, N.; Salin, D. *Phys. Rev. Lett.* **2003**, *90*, 128302.
- Nugent, C. R.; Quarles, W. M.; Solomon, T. H. *Phys. Rev. Lett.* **2004**, *93*, 218301.
- Paoletti, M. S.; Solomon, T. H. *Europhys. Lett.* **2005**, *69*, 819.
- Chemical Waves and Patterns*; Kapral, R.; Showalter, K., Eds.; Kluwer Academic Publishers: Dordrecht, 1995.
- Zaikin, N.; Zhabotinsky, A. M. *Nature* **1970**, *225*, 535.
- Manz, N.; Ginn, B. T.; Steinbock, O. *Phys. Rev. E* **2006**, *73*, 66218.
- Spatio-Temporal Pattern Formation*; Walgraef, D., Ed.; Springer: New York, 1997.
- Nonlinear Evolution of Spatio-Temporal Structures in Dissipative Continuous Systems*; Busse, F. H.; Kramer, L., Eds.; Plenum Press: New York, 1990.
- Li, Y. J.; Oslonovitch, J.; Mazouz, N.; Plenge, F.; Krischer, K.; Ertl, G. *Science* **2001**, *291*, 2395.
- Nonlinear Spatio-Temporal Dynamics and Chaos in Semiconductors*; Scholl, E., Ed.; Cambridge University Press: Cambridge, 2001.
- Ackemann, T.; Lange, W. *Appl. Phys. B* **2001**, *72*, 21.
- Umbanhowar, P. B.; Melo, F.; Swinney, H. L. *Nature* **1996**, *382*, 793.
- Winfrey, T. J. *Biosci.* **2002**, *27*, 465.
- Dahlem, M. A., Ph.D. thesis; Otto-von-Guericke-Universität Magdeburg, 2000.
- Biochemical Oscillations and Cellular Rhythms*; Goldbeter, A., Ed.; Cambridge University Press: Cambridge, 1996.
- The Self-Made Tapestry: Pattern Formation in Nature*; Ball, P., Ed.; Oxford University Press: Oxford, 1999.
- Steinbock, O.; Müller, S. C. Z. *Naturforsch. C* **1995**, *50*, 275.
- Mathematical Biology*; Murray, J. D., Ed.; Springer-Verlag: Berlin, 1989.
- Self-Organized Biological Dynamics & Nonlinear Control*; Walleczek, J., Ed.; Cambridge University Press: Cambridge, 2000.
- Orbán, M. *J. Am. Chem. Soc.* **1980**, *102*, 4311-4314.
- Oscillations and Travelling Waves in Chemical Systems*; Field, R. J.; Burger, M., Eds.; Wiley: New York, 1985.
- An Introduction to Non-linear Chemical Dynamics*; Epstein, I. R.; Pojman, J. A., Eds.; Oxford University Press: New York, 1998.
- Huh, D. S.; Choe, S. J.; Kim, M. S. *React. Kinet. Catal. Lett.* **2001**, *74*, 11.
- Huh, D. S.; Kim, M. S.; Choe, S. J. *Bull. Korean Chem. Soc.* **2001**, *22*, 867.
- Huh, D. S.; Kim, Y. J.; Wang, J. *Phys. Chem. Chem. Phys.* **2003**, *5*, 3188.
- Huh, D. S.; Kim, Y. J.; Choe, S. J. *Bull. Korean Chem. Soc.* **2004**, *25*(2), 267.
- Manz, N.; Ginn, B. T.; Steinbock, O. *Phys. Rev. E* **2006**, *73*, 066218, 1-4.
- Bánsági, T.; Palczewski, C.; Steinbock, O. *J. Phys. Chem. A* **2007**, *111*, 2492-2497.
- Agladze, K.; Thouvenel-Romans, S.; Steinbock, O. *Phys. Chem. Chem. Phys.* **2001**, *3*, 1326-1330.
- Hamik, C. T.; Manz, N.; Steinbock, O. *J. Phys. Chem. A* **2001**, *105*, 6144-6153.
- Diewald, M.; Brand, H. R. *Phys. Rev. E* **1995**, *51*, R5200.
- Hydrodynamic and Hydromagnetic Stability*; Chandrasekhar, S., Ed.; Oxford University Press: London, 1961.