Chemical Waves

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Spatial structures may occur in nonlinear systems far from equilibrium. Chemical waves, which are concentration variations of chemical species propagating in a system, are an example of such structures. A survey is given of some experiments on chemical waves by spectroscopic and microphotographic techniques, arranged according to different types of waves, different geometries, and various properties.

CHEMICAL FRONTS AND WAVES (*1*) ARE VARIATIONS IN concentrations of chemical species, or possibly in other state variables such as temperature, which travel in space and occur in nonlinear reactive systems far from equilibrium. Waves in the so-called Belousov-Zhabotinsky (*2–8*) reaction were first reported by Zhabotinsky (*9*). The overall reaction is the oxidative bromination by acidic bromate of an organic substrate such as malonic acid; it can be approximated by

$$\begin{aligned} 2BrO_3^- + 3CH_2(COOH)_2 + 2H^+ &\rightarrow \\ 2BrCH(COOH)_2 + 3CO_2 + 4H_2O & \qquad (1) \end{aligned}$$

The reaction is catalyzed by an oxidation-reduction couple such as Ce^{3+}/Ce^{4+} or ferroin-ferriin, $Fe(phen)_3{}^{2+}/Fe(phen)_3{}^{3+}$, where phen is phenanthroline. The reaction can be oscillatory, in which case the concentrations of some chemical intermediates vary in time periodically. The period of such oscillations is typically about 40 seconds, and the concentration of Br^- may vary by over five orders of magnitude.

There are many reports of visual observations of waves in this and a few other chemical systems [see (*7, 8*) and citations therein]. Detailed studies by spectroscopic and photographic methods of different types of waves, the structure of the fronts of waves, and other properties such as dispersion relations have appeared only in the last 2 years (*10–19*). In this article we present a brief survey of such experiments on chemical waves considered as reaction-diffusion processes but largely omit theory, calculations, and the interaction of such processes with convection. We begin with a categorization of waves: kinematic, trigger, and phase diffusion waves. We then discuss different geometries: plain, circular, spiral, multiarmed vortices, and scroll waves. Then we consider different properties of waves: the amplitude, the velocity, the front structure, dispersion relations, and the relation of curvature and velocity.

Chemical waves are described by solutions of reaction-diffusion equations

$$\frac{\partial \Psi}{\partial t} = D\nabla^2\Psi + F[\Psi] \qquad (2)$$

where Ψ is a vector of state variables, such as the concentrations of chemical species and possibly temperature, each dependent on space and time; D is a matrix of transport (diffusion) coefficients, each assumed to be constant; and $F[\Psi]$ represents the variations in time that arise from the chemical reactions. These nonlinear equations can seldom be solved in closed form; a variety of approximations and numerical techniques lead to useful solutions. Reviews on the theory of reaction-diffusion equations are available (*8, 20–23*).

The field of temporal and spatial structures in nonlinear chemical systems far from equilibrium, such as chemical systems with multiple stationary states, with oscillations in chemical intermediates and products, with chaotic variations of concentrations and other state

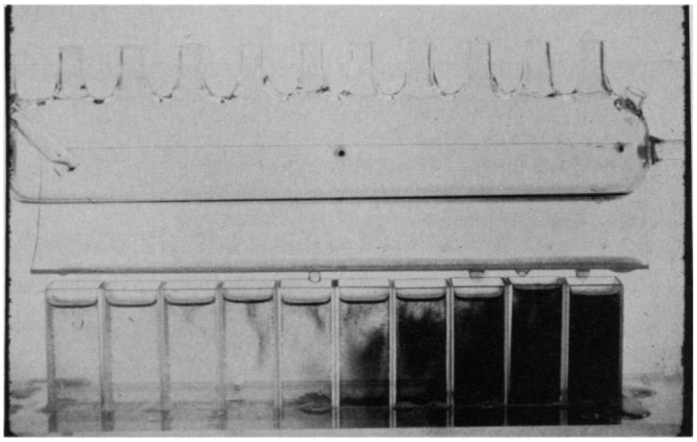

Fig. 1. Kinematic wave in a row of ten adjacent cells. An initial phase (or frequency) gradient imposed on the oscillatory Briggs-Rauscher reaction leads to the apparent propagation of colored fronts. [From (*39*), reprinted with permission, copyright (1982) *Actual. Chim.*]

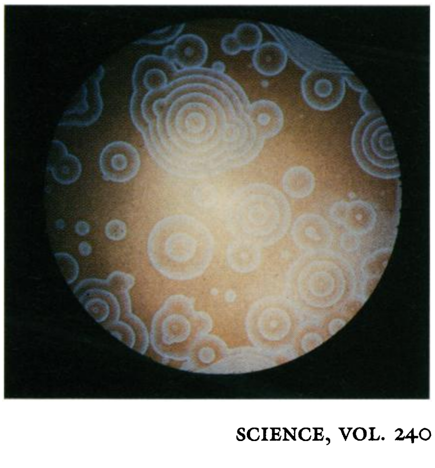

Fig. 2. Two-dimensional target patterns in the Belousov-Zhabotinsky reaction in a layer of solution 1 mm deep. The scale of the photograph is about 5 cm. [From (*13*), reprinted with permission, copyright (1986) *J. Phys. (Paris)*]

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variables, and with spatial structures (*24*) such as chemical waves is receiving significant attention. Systems far from equilibrium are less well understood, both experimentally and theoretically, than systems at equilibrium; yet most natural phenomena are indeed far from equilibrium.

In the past 20 years many examples of oscillatory chemical, biochemical, and biological (both in vivo and in vitro) reactions have been reported (*7, 25*). Waves and fronts in biological systems have been studied for a long time: in signal propagation in nerves (*26–28*); in peristaltic motion (*29*); in the development of embryos (*30, 31*); in phage-bacterium systems (*32*); in the aggregation step of the life cycle of slime mold cells (*33*); in waves across the chambers of the heart (*34, 35*); in pulses of pheromone emission (odor song) (*36*); and in spreading depression in the cerebral cortex (*37*), among others. All these waves can only be electrochemical in origin, and this fact provides additional motivation for the study of the physical chemistry of chemical waves.

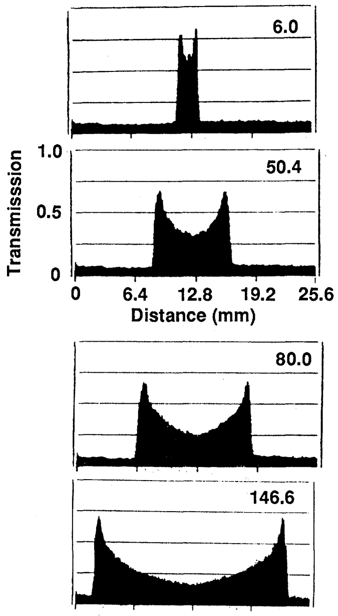
Types of Waves

Kinematic waves. Another oscillatory chemical system is the Briggs-Rauscher reaction (*38*). Consider such a solution placed in the long upper tube shown in Fig. 1 (*39*). On turning the tube so that the many sidetubes face downward, we observe that the vessels below the long tube become filled. If there exists initially either a small variation in frequency of oscillation from one end of the long tube to the other, induced, for example, by a temperature gradient imposed on that tube, or if there exists a phase gradient in the oscillations, then one will see the passage of waves across the separate beakers. Such a wave, which is an optical illusion, is called a kinematic wave (*40, 41*). It clearly does not involve mass transfer from one beaker to another. If an initial phase gradient or frequency gradient is set up in the long tube, then kinematic waves will also sweep from one end of the long tube to the other, initially without mass transfer. However, after a time, owing to concentration differences in neighboring spatial elements of the tube along its axis, diffusion will occur and different types of waves will appear.

Trigger waves. Let us return to the Belousov-Zhabotinsky reaction in an oscillatory state (*42–49*), say, a solution of that reaction in a shallow (1 mm or less) layer in a petri dish. The solution is first well stirred and then left quiescent. Homogeneous concentration oscillations in the ratio of the oxidation reduction couple, say, Fe^{2+}/Fe^{3+}, are observed with an appropriate indicator ferroin [tris(1,10-phenanthroline) ferrous sulfate] [$Fe(phen)_3^{2+}$ is pink, $Fe(phen)_3^{3+}$ is blue]; the color changes from pink to blue and back to pink, repeatedly. After a while inhomogeneities appear, frequently with a gas bubble or dust particle at the center of the inhomogeneity. In time, a chemical front proceeds radially outward from the center of the inhomogeneity. A number of trigger waves are shown in a photograph (*13*) of such a solution in a petri dish (Fig. 2).

The transmission profile of a propagating trigger wave (*10*) in the Belousov-Zhabotinsky reaction in an excitable stationary state is given in Fig. 3. An appropriate perturbation from such a stationary state, of a threshold magnitude, leads to large concentration variations, just as in an oscillation, before return to the stationary state. The measurements were made by shining a diffuse laser beam onto the surface of the solution, perpendicular to the plane of the layer of the solution. The transmitted light is measured on a diode array system which consists of 1024 photosensitive spots located on a line 25 μm apart. The array can be scanned about once a second, and the data are stored in a computer system and displayed on a screen. By repeating such measurements in time we obtain the concentration of Fe^{2+} (the dominant absorbing species) as a function of space and time and hence a quantitative analysis of the front as it propagates in space and time.

Fig. 3. Wave propagation sequence from initiation at a single point; transmission of argon ion laser pulse at 488 nm as a function of distance. Temperature is 18.0°C. Time (in seconds) since wave initiation is shown in the upper right corner of each scan. [From (*10*), reprinted with permission, copyright (1985) American Institute of Physics]



Trigger waves have sharp fronts, 0.1 to 0.5 mm in width [concentration gradients up to 17 m*M*/mm of ferroin (*18*)] that arise from a phase in the oscillatory Belousov-Zhabotinsky reaction during which rates are very rapid and hence concentration variations are steep in time. Both reaction and diffusion play crucial roles in the propagation of trigger waves. Trigger waves do not interpenetrate each other but annihilate each other on contact, when concentration gradients are equal and opposite in the two colliding waves; unlike kinematic waves, they are blocked by walls.

Trigger waves in an excitable medium are induced by a disturbance of the right direction and magnitude at one location, which leads to sharp concentration differences (gradients) with neighboring locations. Diffusion occurs, which then sets off the same disturbance in the neighboring locations with consequent propagation of a front. The mechanism of wave propagation in an oscillatory reaction is the same.

Phase diffusion waves. Consider a solution of the Belousov-Zhabotinsky reaction, in a petri dish, which oscillates autonomously. We impose a well-focused laser beam, of wavelength absorbed by Fe^{2+}, on a small area (0.2 mm^2) and irradiate the solution for a limited time (about 12 seconds). The photons absorbed are converted into heat, which raises the temperature of the volume beneath the small area and brings about a perturbation that sets off a trigger wave (see Fig. 4, first cycle, graphs at 108.1 to 113.3 seconds) (*11*). That wave, with sharp concentration fronts, propagates into the unperturbed medium. When the phase of the oscillation of the concentration of Fe^{2+} in the unperturbed medium matches that of the wave front, the front disappears (Fig. 4, first cycle, graph at 130.3 seconds). However, because of the temperature difference between the irradiated spot and the remainder of the solution, there exists a difference in frequency of oscillation and after a passage of a cycle the wave reappears. With the elapse of some time, the energy deposited by the light, turned into heat in the solution, diffuses and the temperature gradient becomes shallower. As that happens, a different wave appears (Fig. 4, fourth cycle, graphs at 449.0 to 453.0 seconds), called a phase diffusion wave (*41, 50*). The velocity of a phase diffusion wave is defined as the ratio of the variation of the phase with time divided by the variation of the phase in space

$$v = -\frac{(\partial\phi/\partial t)}{(\partial\phi/\partial r)} \qquad (3)$$

Hence, as the heat in the solution at the irradiated spot diffuses away, the phase gradient in space becomes smaller and hence the velocity of the phase wave increases. When the velocity of the phase diffusion wave exceeds that of the trigger wave (of essentially constant velocity), then a phase wave and not a trigger wave propagates in the solution. Velocities of phase diffusion waves are nearly constant for a limited time interval and become very large as the phase difference between the wave and the solution disappears (see Eq. 3). The concentration gradients in a phase diffusion wave are much shallower than those in a trigger wave; compare, for instance, the gradients at 451.6 seconds with those at 110.7 seconds in Fig. 4. In an autonomous oscillatory reaction, phase diffusion waves appear only if concentration gradients are small; for large concentration gradients, trigger waves appear.

Fronts in bistable systems. Chemical reactions far from equilibrium may have multiple, stable stationary states. On transition from one such stable state to another, a front of concentration variations travels through space. Such fronts have been observed in the iodate–arsenous acid reaction (*51*) and others.

Geometric Forms of Waves

The fronts of waves of chemical activity may assume various shapes depending on the geometry and volume of the container. All the characteristic features of these shapes are best realized in quiescent solutions in which trigger waves can be excited.

One-dimensional waves. These waves are formed in a confined space, as in a narrow (a few millimeters) test tube, and consist of a train of fronts following each other at a distance. Measurements in such simple geometries are performed, for instance, in order to study the effect of an externally applied electrical field on wave properties (*52, 53*). The dependence of the velocity of chemical waves on electric field intensity is nonlinear. Waves can be slowed with appropriate polarity of the field; at high fields, waves may be split and the direction of propagation may be altered.

Two-dimensional waves. In a thin layer (about 1 mm^2) of reactive solution in a petri dish there occur several types of two-dimensional wave forms. An apparatus (*14–19*) for computerized digital spectrophotometry of structures in two dimensions consists of ultraviolet (UV) optical components mounted on a vibration-isolated table for illumination and imaging purposes, a UV-sensitive video camera serving as the two-dimensional intensity detector, and a fast, large-memory computer for storage of the digitized data and further data processing. The sample layer in an optically flat petri dish is illuminated from above with a parallel, spatially homogeneous light beam emerging from a 300-W xenon short-arc lamp (Cermax) that has high temporal stability. Square sections (1 by 1 mm^2 to 15 by 15 mm^2) are imaged by a UV photo lens on the target of the video camera with an image raster resolution of 512 by 512 picture elements (pixels). The video signal is converted to digital data with 256 digital units (gray levels) intensity resolution. The apparatus combines spatial, temporal, and intensity resolution satisfactory for the analysis of chemical patterns and their temporal evolution. A comprehensive software package for the presentation of two-dimensional data arrays includes extraction of profiles of transmitted light intensity, logarithmic conversion of intensities into concentrations, pseudocolor and three-dimensional perspective graphical presentations, and fitting procedures for specific isointensity or isoconcentration lines.

1) Distributed sets of concentric annuli (*42–49*) are frequently also called "target" patterns (Fig. 2) and have been discussed in the section on trigger waves. One important property of these waves is their mutual annihilation upon collision, which leads to typical cusplike structures in the vicinity of the area of collision between two annuli.

2) Spiral-shaped waves are formed by the disruption of an expanding circular wave front, as can be done in a controlled manner with a gentle blast of air from a pipette onto the surface of the reacting solution (*15, 43, 45*). The irregularly shaped open ends of the circular wave are then the starting points of a rapid evolution toward a structure composed of a pair of counterrotating spiral waves with highly regular geometry. The tips of these spiral-shaped vortices turn inward with a rotation period of 17 seconds, whereas the fronts move in the outward direction. Spiral waves have been studied in some detail (*14–17*). Figure 5A shows the digital image of a pair of spiral waves in three-dimensional perspective presentations with a specific isoconcentration level of the light-absorbing catalyst marked in black. Outside a small region surrounding the center of rotation, the so-called spiral core, the structure follows in good approximation an Archimedean geometry, but the involute of a circle fits the structure equally well. These two curves are asymptotically identical and differ only slightly in the immediate neighborhood of the core, where the resolution of the measured data points is not yet sufficient to distinguish between them. Theoretical analysis of simplified models of this reaction yielded results close to the shape of the involute (*54*). The three-dimensional

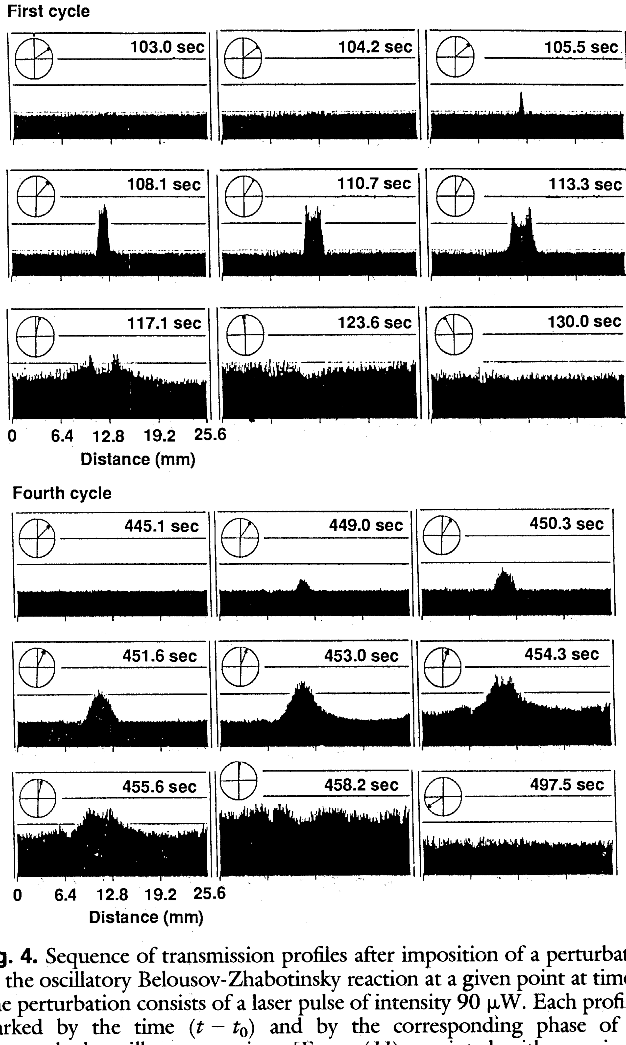


Fig. 4. Sequence of transmission profiles after imposition of a perturbation on the oscillatory Belousov-Zhabotinsky reaction at a given point at time t_0. The perturbation consists of a laser pulse of intensity 90 μW. Each profile is marked by the time $(t - t_0)$ and by the corresponding phase of the unperturbed oscillatory reaction. [From (*11*) reprinted with permission, copyright (1987) American Institute of Physics]

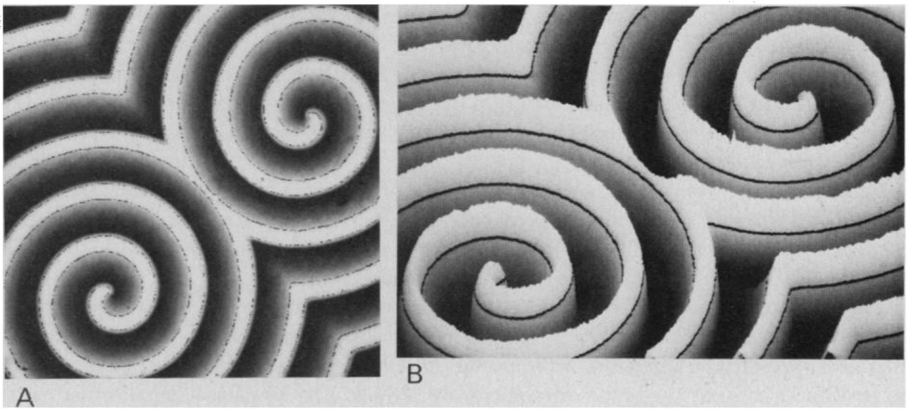


Fig. 5. (**A**) Symmetric pair of counterrotating spiral waves in a thin layer of an excitable Belousov-Zhabotinsky reaction. The digital image shows an 8.2 mm by 8.2 mm area and is composed of 410 × 410 picture elements. Each element has one out of 256 possible gray levels of transmitted light intensity, which is a measure of the local concentration of the catalyst ferroin. One concentration level is enhanced in black. (**B**) The inner section of (A) is rendered as a three-dimensional surface image by interpolation of the measured pixel values and subsequent projection on perspective. The level enhanced in black is the same as that in (A). [From (*18*), reprinted with permission, copyright (1987) *Biophys. Chem.*]

perspective image in Fig. 5B indicates the asymmetric shape of the wave profiles. The gradients of ferroin are about 10 m*M*/mm at the steep fronts and 1 m*M*/mm at the gentler sloping backs. The rotation center is a singular site in that it is the only location in the system where the chemistry remains quasi-stationary; in the areas outside the center periodic redox transitions take place that are correlated with the outward propagation of the wave front. The size of the site is smaller than 10 μm. The core region is just that portion of the layer where a transition takes place from the spiral center to the outer area where the solution is excited to full-amplitude waves. This transition gives the core a "tornado"-like structure, as shown in the cover.

3) Multiarmed vortices have been seen in a shallow layer of an excitable medium. They are obtained by the controlled addition of a drop of a chemical close to the center of rotation of a spiral wave. Two-, three-, and four-armed vortices, that is, spiral waves having the same center of rotation, have been produced (*55*).

Three-dimensional waves. Evidence for three-dimensional wave spheres first came from experiments with stacks of filter paper (*45*). Later such waves were observed in a liquid medium contained in a cylinder (8 mm), whereby special care was taken to avoid disturbances arising from bubbles and fluid motion. Predominant wave types are toroidal scroll waves, but occasionally other types such as spheroidal waves are observed (*56*). Analytical and numerical predictions have been made for the possibility of twisted and linked scroll rings (*57*). Their experimental realization, however, as well as the quantitative investigation of any three-dimensional structure remains a challenge for future work.

All these experiments to produce different forms of waves are carried out in closed (batch) systems and not in the open reactors usually used for the investigation of oscillatory behavior in homogeneous systems. Thus the reagent mixture undergoes aging, which results in a slow drift of the reacting system toward equilibrium. A first approach to experimentation on chemical waves in an open system has been reported; sustained wave patterns were realized in an open annular reactor (*58*). The structure consists of traveling azimuthal wave pieces which look very much like pinwheels.

Properties of Chemical Waves

Shape and amplitude. If we select a wave with a single bright (blue-oxidizing) wave front and measure the transmission profile along a diameter of the circular wave, then the results are as shown in Fig. 3 for a trigger wave in the Belousov-Zhabotinsky reaction in an excitable stationary state. A three-dimensional representation of photographs of the same reaction in oscillatory conditions is shown in Fig. 6. The photographs are subjected to a quantitative analysis by first recording the images from a TV camera on video tape and then digitizing selected frames of pictures. With the techniques of geometrical corrections, background subtraction, enhancement of pattern by filtering and thresholding, among others, characteristic features of the patterns can be obtained. The oxidation domain grows from a small area around the center. A flat cylinder appears with radius and height (amplitude) increasing monotonically. Beyond a critical value of the radius, the reduction phase then begins at the center and the amplitude of the cylinder at the center decreases.

The constancy of the front and shape of a trigger wave (*10*) is shown in Fig. 7. Figure 7A gives a measurement of the transmission profile of Fe^{2+} in space at a given time. Figure 7B is the transmission profile at a given point in space as a function of time. Figure 7C shows a superposition of the measurements in (A) and (B) translated by the measured velocity of propagation of the front according to the equation $\phi = X - vt$, where ϕ is the phase, X is position, and t is time. The constancy of the shape in time is quantitatively confirmed. A wave initiated in water when a rock is dropped into it propagates with constantly decreasing amplitude due to the viscosity of the medium. The comparison, however, is not fair since the water wave propagates into a medium at equilibrium. In the case of chemical trigger waves the disturbance propagates into a medium that is far from equilibrium. In spite of the fact that diffusion removes concentration gradients, the combination of reaction and diffusion propagates the wave with constant shape. The energy necessary for this process comes from the Gibbs free energy change of the reaction. With respect to the constancy of wave profile, energetics, and nonlinear origin, there are interesting similarities between these waves and nerve conduction; the velocity of chemical waves,

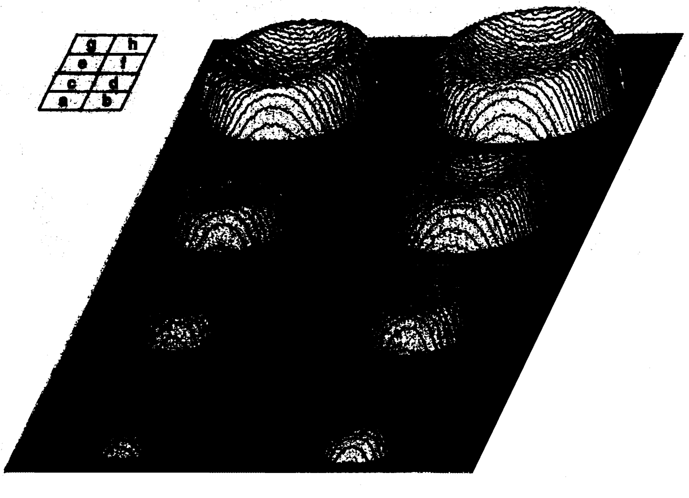


Fig. 6. Three-dimensional perspective representation of light transmission by a layer of oscillatory Belousov-Zhabotinsky reaction. The eight successive snapshots, taken at 1-second intervals, show the buildup of the oxidizing wave front. [Courtesy of C. Vidal and A. Pagola, in preparation]

however, is considerably slower (by about a factor of 400,000).

The range of variation in the concentration of Fe^{3+} in an oscillation is a factor of 2 or 3 (from $1 \times 10^{-4}M$ to $3 \times 10^{-4}M$). The distance in which this sharp variation occurs, the front width, varies with initial concentration in the system in a manner that has not yet been explored.

Velocity. At a given temperature the velocity of a trigger wave depends on the initial concentrations of reactants, 4.0 mm/min for the wave shown in Fig. 3. The velocity has a square root dependence (*10, 46, 47*) on the concentrations of sulfuric acid and bromate ion, $v \sim k[H_2SO_4]^{1/2}[BrO_3^-]^{1/2}$, in a limited range of initial concentrations, and is much less sensitive to the concentrations of the other chemical species. The variation of the coefficient k has been studied so far only over a narrow range of temperature (284 to 318 K) and is reasonably given by an Arrhenius law with an apparent activation energy of about 35 kJ mol^{-1}.

Dispersion of chemical waves. Dispersion is the variation of the velocity of wave propagation with the period (of oscillation in an oscillatory chemical system). Consider the emission of a wave from a center with a period T into a medium of oscillatory period T_0. The first wave emitted from the center propagates into the medium with a velocity that is described by the concentrations in the medium. If the period of emission is of the same order as T_0, then the second and successive waves propagate into a medium that is essentially relaxed after the passage of the prior wave, and the velocity of the propagation is little affected. However, if the period of emission is below $T_0/2$, then successive waves propagate into a medium that is incompletely relaxed and a reduction in the velocity of propagation may be expected. Measurements of the dispersion relation (*12*) in an oscillatory Belousov-Zhabotinsky reaction show that for T/T_0 larger than 0.5, the reduction in velocity in propagation is small, whereas for T/T_0 smaller than 0.5 the reduction in propagation velocity is substantial.

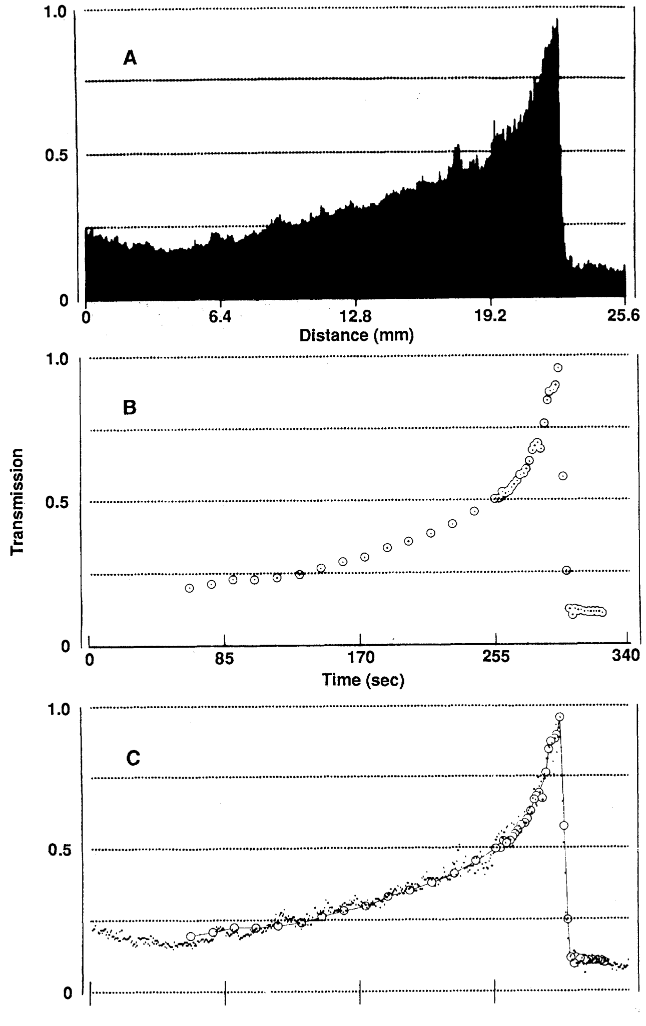


Fig. 7. Transmission profile of a trigger wave: (**A**) sequence similar to Fig. 3, as a function of distance on a reticon; (**B**) sequence as a function of time on one photosensitive spot on the reticon; (**C**) superposition of the measurements in (A) and (B) as described in text. The superposition shows the constancy of the wave profile in time. [From (*10*), reprinted with permission, copyright (1985) American Institute of Physics]

In order to solve reaction-diffusion equations numerically, we must have a reaction mechanism and rate coefficients for the various steps in that mechanism. Much effort has gone into unraveling the mechanism of complex reactions, in particular that of the Belousov-Zhabotinsky reaction, and a number of simplified models of the mechanism have been proposed. Even a two-variable model (*54, 59–61*), which oversimplifies the mechanism and neglects diffusion of all but one of the reacting species (bromous acid), provides a fairly good description of the main features of the wave profile, the velocity of propagation, and dispersion.

Origin of trigger waves. A fundamental point, not yet settled, concerns the origin of trigger waves that appear spontaneously in a thin layer of solution of an oscillatory or excitable chemical system. Do these waves arise spontaneously (*62*) as a result of a symmetry-breaking fluctuation? Or do these waves arise deterministically from a heterogeneous center (a dust particle, an impurity, a gaseous bubble)? Extensive measurements have been made (*13*) on samples of more than 500 centers, and distributions of velocities of wave propagation have been determined, with no conclusive decision as yet. Experiments on thoroughly filtered solutions of the Belousov-Zhabotinsky reaction show that formation of trigger waves can thus be suppressed in an excitable system but not in an oscillatory system. A theory has been proposed for these results, which favors heterogeneous centers (*61*).

Propagation velocity and curvature in spiral waves. An important problem in the study of spiral waves is the relation between the curvature and the propagation velocity of a front (*63*). Quantitative confirmation of the curvature-velocity equation used by Keener and Tyson (*54*) has been obtained (*64*) by measuring the temporal evolution of the cusplike structures that form immediately after wave collision (compare the dip in the wave crests in Fig. 5B). Here one can easily produce areas of extremely high curvature and follow the very rapid changes with a video movie.

Conclusions

Chemical waves are an interesting phenomenon that characterizes many nonlinear reaction systems far from equilibrium. The quantitative investigation of these waves is in the initial stage. Much work needs to be done including the study of their characteristics in the limit of zero velocity, that is, stable spatial (Turing) structures, and their application to biological systems.

REFERENCES AND NOTES

1. The earliest report of a chemical wave appears to be that of R. Luther [*Z. Elektrochem.* 12, 596 (1906)].
2. B. P. Belousov, in *Sb. Ref. Radiat. Med.* (collection of abstracts on radiation medicine) (Medgiz, Moscow, 1959), p. 145; A. M. Zhabotinsky, *Biofizika* 9, 306 (1964).
3. For references on this and other oscillatory reactions, see (*4–8*).
4. R. M. Noyes and R. J. Field, *Annu. Rev. Phys. Chem.* 25, 95 (1974).
5. J. J. Tyson, *The Belousov-Zhabotinsky Reaction*, Lecture Notes on Biomathematics, (Springer-Verlag, Berlin, 1976), vol. 10.
6. C. Vidal and A. Pacault, in *Evolution of Order and Chaos*, H. Haken, Ed. (Springer-Verlag, Heidelberg, 1982), p. 74.

7. R. Field and M. Burger, Eds., *Oscillations and Traveling Waves in Chemical Systems* (Wiley, New York, 1985).
8. C. Vidal and P. Hanusse, *Int. Rev. Phys. Chem.* **5**, 1 (1986).
9. A. M. Zhabotinsky, in *Oscillatory Processes in Biological and Chemical Systems*, G. M. Frank, Ed. (Science Publishers, Moscow, 1967), p. 252.
10. P. M. Wood and J. Ross, *J. Chem. Phys.* **82**, 1924 (1985).
11. J. M. Bodet, C. Vidal, J. Ross, *ibid.* **86**, 4418 (1987).
12. A. Pagola, C. Vidal, J. Ross, *J. Phys. Chem.* **92**, 163 (1988).
13. C. Vidal *et al.*, *J. Phys. (Paris)* **47**, 1999 (1986).
14. S. C. Müller, Th. Plesser, B. Hess, *Naturwissenschaften* **73**, 165 (1986).
15. ———, *Science* **230**, 661 (1985).
16. ———, *Physica* **24D**, 71 (1987).
17. ———, *ibid.*, p. 87.
18. ———, *Biophys. Chem.* **16**, 357 (1987).
19. Zs. Nagy-Ungvarai *et al.*, *Naturwissenschaften*, in press.
20. H. Eyring and D. Henderson, Eds., *Theoretical Chemistry, Advances and Perspectives*, vol. 4, *Periodicities in Chemistry and Biology* (Academic Press, New York, 1978).
21. P. Fife, *Mathematical Aspects of Reacting and Diffusing Systems* (Springer-Verlag, New York, 1979).
22. Y. Kuramoto, *Chemical Oscillations, Waves, and Turbulence* (Springer-Verlag, Heidelberg, 1984).
23. P. Hanusse, J. Ross, P. Ortoleva, *Adv. Chem. Phys.* **38**, 317 (1978).
24. A. M. Turing, *Philos. Trans. R. Soc. London Ser. B* **237**, 37 (1952).
25. B. Hess and A. Boiteus, *Annu. Rev. Biochem.* **40**, 237 (1971); P. E. Rapp, *J. Exp. Biol.* **81**, 281 (1979).
26. R. Fitzhugh, *Biophys. J.* **1**, 445 (1961).
27. E. C. Zeeman, in *Towards a Theoretical Biology* (Aldine-Atherton, Chicago, 1972), vol. 4, p. 8.
28. G. Matsumoto and H. Shimizu, *J. Phys. Soc. Jpn.* **44**, 1399 (1978).
29. N. W. Weisbrodt, in *Physiology of the Gastrointestinal Tract*, L. R. Johnson, Ed. (Raven, New York, 1981), vol. 1, pp. 411–433.
30. L. Wolpert, *J. Theor. Biol.* **25**, 1 (1969).
31. A. Gierer and H. Meinhardt, *Kybernetik* **12**, 80 (1972).
32. G. R. Ivanitsky, A. S. Kunisky, M. A. Tzyganov, in *Self-Organization—Autowaves and Structures Far from Equilibrium*, V. I. Krinsky, Ed., vol. 28 of Synergetics Series (Springer-Verlag, Berlin, 1984), p. 214.
33. G. Gerisch, *Curr. Top. Dev. Biol.* **3**, 157 (1968); J. L. Martiel and A. Goldbeter, *J. Biophysics* **52**, 807 (1987), and references therein.
34. M. A. Allessie, F. I. M. Bonke, F. J. G. Schopman, *Circ. Res.* **41**, 9 (1977).
35. A. T. Winfree, *Sci. Am.* **248** (no. 5), 144 (1983).
36. W. H. Bossert, *J. Theor. Biol.* **18**, 157 (1968).
37. J. Bures, V. I. Koroleva, N. A. Gorelov, in (*32*), p. 180.
38. T. S. Briggs and W. C. Rauscher, *J. Chem. Educ.* **50**, 496 (1973).
39. M. Sadoun-Goupil, P. de Kepper, A. Pacault, C. Vidal, *Actual. Chim.* 7, 37 (1982).
40. N. Koppel and L. N. Howard, *Science* **180**, 1171 (1973).
41. P. Ortoleva and J. Ross, *J. Chem. Phys.* **60**, 5090 (1974).
42. A. N. Zaikin and A. M. Zhabotinsky, *Nature (London)* **225**, 535 (1970).
43. A. T. Winfree, *Science* **175**, 634 (1972).
44. A. M. Zhabotinsky and A. N. Zaikin, *J. Theor. Biol.* **40**, 45 (1973).
45. A. T. Winfree, *Sci. Am.* **230** (no. 6), 82 (1974).
46. R. J. Field and R. M. Noyes, *J. Am. Chem. Soc.* **96**, 2001 (1974).
47. T. A. Gribschaw, K. Showalter, D. L. Banville, I. R. Epstein, *J. Phys. Chem.* **85**, 2152 (1981); L. Kuhnert, H.-J. Kong, L. Pohlman, *J. Phys. Chem.* **89**, 2022 (1985).
48. M. Orban, *J. Am. Chem. Soc.* **102**, 4311 (1980).
49. P. De Kepper, I. R. Epstein, K. Kustin, M. Orban, *J. Phys. Chem.* **86**, 170 (1982).
50. There is no unaminity on nomenclature. What we call kinematic waves are sometimes referred to as phase waves.
51. J. Harrison and K. Showalter, *ibid.* **90**, 225 (1986).
52. P. J. Ortoleva and S. L. Schmidt, in (*7*), p. 333.
53. H. Szevcikova and M. Marek, *Physica* **13D**, 379 (1984).
54. J. P. Keener and J. J. Tyson, *ibid.* **21D**, 307 (1987).
55. K. I. Agladze and V. I. Krinsky, *Nature (London)* **296**, 424 (1982).
56. B. J. Welsh, J. Gomatam, A. E. Burgess, *ibid.* **304**, 611 (1983).
57. A. T. Winfree and S. H. Strogatz, *Physica* **13D**, 221 (1983).
58. Z. Noszticzius, W. Horsthemke, W. D. McCormick, H. L. Swinney, W. Y. Tam, *Nature (London)* **329**, 619 (1987).
59. J. D. Dockery, J. P. Keener, J. J. Tyson, in preparation.
60. J. J. Tyson and P. Fife, *J. Chem. Phys.* **73**, 2224 (1980).
61. J. J. Tyson, *ibid.*, in press.
62. D. Walgraef, G. Dewel, P. Borckmans, *ibid.* **78**, 3043 (1983).
63. V. S. Zykov and G. L. Morozova, *Biofizika* **23**, 717 (1979).
64. P. Foerster, S. C. Müller, B. Hess, in *Spatial Inhomogeneities and Transient Behavior in Chemical Kinetics*, G. Nicolis and P. Gray, Eds. (Springer-Verlag, Berlin, in press).

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