Reports

The Motion of Untwisted Untorted Scroll Waves in Belousov-Zhabotinsky Reagent

JAMES P. KEENER AND JOHN J. TYSON

Rotating waves of activity are seen in various biological phenomena and in chemical mixtures. In thin layers of these media, the waves often appear as spirals spinning around a pivot point, but actually they are scroll-shaped waves rotating around a curved filament in three-space. The filament about which the scroll rotates is not stationary, but rather moves through space until it achieves a stable configuration or disappears altogether. Some features of the temporal evolution of a planar scroll wave filament can be understood in terms of the simple equation $N = D\kappa$, where N is the velocity of the filament in the direction of its principal normal, κ is the curvature of the filament, and D is the diffusion coefficient of the active chemical species. This equation of motion implies that a scroll ring shrinks in size and collapses in finite time, that an elongated spiral evolves into a symmetric spiral, and that an elongated target pattern becomes more symmetrical and vanishes in finite time. Characteristic times for these processes are estimated. In each case, good quantitative agreement is found between implications of the model and observations of scroll-wave evolution in shallow layers of the Belousov-Zhabotinsky reagent.

ROTATING WAVES OF ACTIVITY ARE observed in chemical systems, such as Belousov-Zhabotinsky (BZ) reaction (*1*), and in biological systems, such as slime mold aggregation (*2*), heart muscle contraction (*3*), and cerebral cortex activity (*4*). Because of experimental difficulties involved in studying rotating waves in cellular tissues, the BZ reaction has proved extremely useful. Under appropriate conditions, the BZ reaction supports rotating scroll waves of oxidation (*5–7*). The scroll rotates around a central filament that topologically is a one-dimensional locus of phase singularity (*8*). The filament threads through the three-dimensional medium until it joins to itself, or terminates at a boundary. Depending upon the shape of the filament, the scroll wave manifests itself in one of several forms: symmetric spiral, elongated spiral, elongated target pattern, scroll ring, and (possibly) scroll knot (*5, 8, 9*).

A shallow layer of BZ reagent, like heart wall or cerebral cortex, is extensive in two spatial directions and limited, but not negligible, in thickness. In such contexts, the upper and lower boundaries of the excitable domain play significant roles in the evolution of scroll waves. To show this, we derive heuristically an equation of motion for the filament of a scroll wave, solve the equation in some simple cases with and without boundary effects, and compare our solutions with experimental observations of evolving scroll waves in BZ reagent.

The topology of a scroll wave can be described by three parameters along the scroll wave filament in three-space: the curvature (κ) and torsion (τ) of the filament and the twist (ϕ_s) of the wave around the filament (*10*). In this report we consider only the effects of filament curvature on the temporal evolution of untwisted ($\phi_s = 0$), untorted ($\tau = 0$) scroll waves. In Fig. 1 a small segment of an untwisted untorted scroll wave is shown in the tangent plane of the filament. The filament, with curvature κ, sheds activity waves first to one side and then to the other as the scroll wave rotates. According to the eikonal equation for wave propagation in excitable media (*11, 12*), the activity waves propagate to one side with speed $N = c + D\kappa$ and to the other side (in the opposite direction) with speed $N = c - D\kappa$. (Here, c is the speed of propagation of plane waves in the excitable medium.) It seems natural to propose that the filament moves normal to itself at speed $N = D\kappa$, the average of the speeds of the waves propagating to each side.

More formal derivations of $N = D\kappa$ have been given in other contexts (*13, 14*). In a general treatment of scroll wave evolution (*14*), $N = D\kappa$ is shown to be the correct equation of motion for a planar curve as long as all state variables have the same diffusion coefficients, but not correct for scrolls with torsion, twist, or unequal diffusion coefficients. We propose to examine the equation $N = D\kappa$ in some detail to see how it accounts in simple fashion for some features of the evolution of untwisted untorted scroll waves.

The equation $N = D\kappa$ is well known to differential geometers and has been studied extensively for curves in the plane and surfaces in three-space (*15*). Any simple closed planar curve obeying $N = D\kappa$ shrinks and asymptotically approaches a circle. Thus it is reasonable to assume that the filament of a scroll ring is a circle. For a circle (*13, 16*), the equation of motion simplifies to $dr/dt = -D/r$, and the solution is easily seen to be

$$r(t) = (r_0^2 - 2Dt)^{1/2} \qquad (1)$$

The ring vanishes in finite time $r_0^2/2D$. Furthermore, the radius changes relatively slowly at first, but collapses dramatically at its death (see Fig. 2). An appropriate diffusion coefficient for BZ reagent is $D = 0.12$ mm^2/min, so the lifetime of a modest scroll ring ($r_0 = 3$ mm, for example) should be about 40 minutes. Welsh, Gomatam, and Burgess (*7*) photographed a scroll ring with a radius of about 3 mm that shrank and disappeared after approximately 60 minutes.

In a shallow layer of excitable medium, the filaments of scroll waves rarely form closed rings, but typically intersect the top or bottom surface of the medium. Scroll waves can be truncated in two ways that lead to two distinctly different behaviors. If the filament intersects both boundaries, one will observe from above an elongated spiral wave, whereas if the filament intersects the same boundary at both ends, one will observe from above an elongated target pattern (*5, 9*). Both elongated spirals and target ("hot dog") patterns are observed in BZ reaction (*5, 6, 9*). As time proceeds, hot dog sources shrink and disappear, whereas elon-

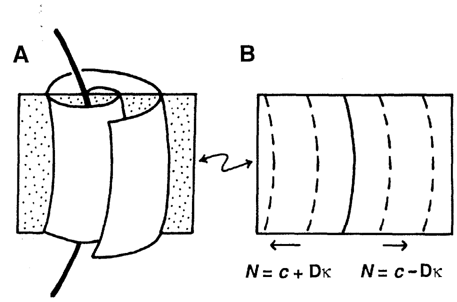

Fig. 1. Motion of the scroll's filament. (**A**) Small section of a scroll wave. The filament of the scroll is the dark curved line at the center of the scroll. The filament is curved but has no torsion. The scroll is not twisted around the filament. The tangent plane to the filament is stippled. (**B**) Activity waves in the tangent plane. As the scroll rotates around its filament, activity waves propagate away from the filament. In the tangent plane, these waves propagate with speed $N = c + D\kappa$ to one side and with speed $N = c - D\kappa$ to the other. The figure suggests that the filament itself will move in its tangent plane at speed $N = D\kappa$.

J. P. Keener, Department of Mathematics, University of Utah, Salt Lake City, UT 84112.
J. J. Tyson, Department of Biology, Virginia Polytechnic Institute and State University, Blacksburg, VA 24061.

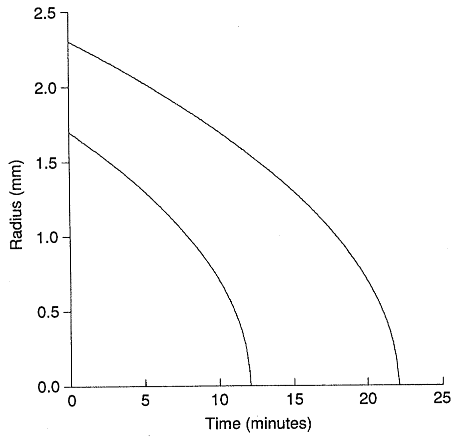

Fig. 2. Life history of scroll rings. The radius of a perfectly circular scroll ring decreases with time according to Eq. 1, with $D = 0.12$ mm^2/min.

gated spirals become symmetric and rotate stably. This difference in behavior can be explained in quantitative detail by examining the equation of motion of the scroll wave filament.

For a filament that intersects both boundaries, we suppose that the filament is a plane curve and its position can be represented by $x = x(z,t)$, where x is the horizontal displacement and z is the depth of a point on the filament. In terms of these variables, the equation of motion of the filament is

$$\partial x/\partial t = D\,\frac{\partial^2 x/\partial z^2}{1 + (\partial x/\partial z)^2} \qquad (2)$$

with boundary conditions $\partial x/\partial z = 0$ at $z = 0, d$. ("No-flux" boundary conditions at $z = 0, d$ are used because in a closed chemical medium there can be no flux of any chemical species across a boundary.) Solutions of Eq. 2 will asymptotically approach a constant that corresponds to a vertical filament for the scroll wave. Thus a scroll wave that intersects both boundaries of the fluid will not collapse, as does a scroll ring, but will approach a stable symmetric shape.

We cannot solve Eq. 2 exactly, but we can approximate its solution readily. If we suppose that the slope of the filament x_z is not too large, and ignore x_z^2 compared to one, then Eq. 2 can be replaced by the heat equation with diffusion coefficient D and no-flux boundary conditions $z = 0, d$. This approximation has the effect of making the diffusion coefficient bigger than it should be, so the resulting estimates of the motion of the filament are too fast. It should, however, be a good approximation in the later stages of the symmetrization of an elongated spiral, when $|x_z| << 1$. Alternately, one could replace the term x_z by its maximal value at time $t = 0$ and obtain an approximate heat equation with diffusion coefficient that is too small, giving estimates of the motion of the filament that are too slow.

Suppose the initial position of the filament is described by the straight line $x(z) = Az$, with $A = L/d$, d = depth of the medium, and L = length of the elongated spiral source projected onto the surface of the medium. The solution of the heat equation with no-flux boundary conditions is given by

$$x(z, t) = L/2 - 4L\pi^{-2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} \exp \left[\frac{-D_m(2n+1)^2\pi^2 t}{d^2}\right] \cos\left[\frac{(2n+1)\pi z}{d}\right] \qquad (3)$$

where D_m is the modified diffusion coefficient $D/(1 + A^2) < D_m < D$. An excellent approximation for Eq. 3 is to keep only the first trigonometric term.

The tilt of the scroll filament is visualized in a shallow layer of BZ reagent as elongation of the spiral wave fronts. From Eq. 3 we estimate the elongation of a tilted scroll as

$$L(t) = x(d, t) - x(0, t) = (8L_0/\pi^2)\exp(-D_m\pi^2 t/d^2) \qquad (4)$$

The tilt of the spiral decays with half-life $T = (d^2 \ln 2)/(\pi^2 D_m)$. For a layer of BZ reagent with depth 1 mm and $D = 0.12$ mm^2/min, an elongated spiral of small tilt ($A < 0.5$, for example) straightens up with a half-life of approximately 1/2 minute. For elongated spirals of greater tilt ($A > 1$), we expect that the half-life of straightening is not much greater than this, because according to Eq. 2 changes in x are greatest where $\partial^2 x/\partial z^2$ is largest, and these are just the places where $\partial x/\partial z$ is relatively small.

In Fig. 3A we show a plot of $L(t)/2$, the half-length of the spiral filament, determined by numerical integration of Eq. 2. As can be seen from these curves, the half-life of shortening is on the order of 1 minute. In Fig. 3B, we plot the same solutions as the $\ln[(L(t)/2]$ versus time. As can be seen from these curves, the solution is sublinear and asymptotically linear. From the slope of the asymptotic linear portion of the curve (-1.2 min^{-1}), we estimated an asymptotic half-life of shortening of 0.6 minute. Using time-lapse photographs of oxidation waves in BZ reagent (*17*), we measured the rate of decay of elongation of four spiral patterns. Because the time-lapse interval was 30 seconds, the accuracy of the measurements was limited, but spirals of length 2 mm or so decayed to perfect symmetry with a half-life of approximately 1 minute. In Fig. 3B the length as a function of time for two of these spirals is recorded.

If the filament of the scroll intersects the same boundary, the scroll wave will disappear in finite time. To see this we suppose that the filament is planar, and solve the equation of motion subject to the requirement that the filament be perpendicular to the boundary at its ends. As a first approximation we could suppose that the filament is semicircular, and apply the estimate for the collapse of full spiral rings found above. This estimate is too generous. To do better, we use a comparison theorem (a noncrossing theorem) for the equation of motion, which states that any two solutions of the equation that are in the same plane and do not cross at time $t = 0$ do not cross for all subsequent times. Furthermore, any planar curve moving with normal velocity that is nowhere larger than D times its curvature cannot cross a solution of the equation of motion if the true solution curve lies inside the comparison curve initially.

Suppose that at time $t = 0$ the filament lies below some straight line with quarter-circular ends (Fig. 4). The radius r_0 of each quarter-circular end is the same as the height of the straight line above $z = 0$, say $r_0 = z_0$. If we allow the quarter-circular ends to move toward each other at speed D/r_0, and the height of the straight line to remain unchanged, then any solution lying under

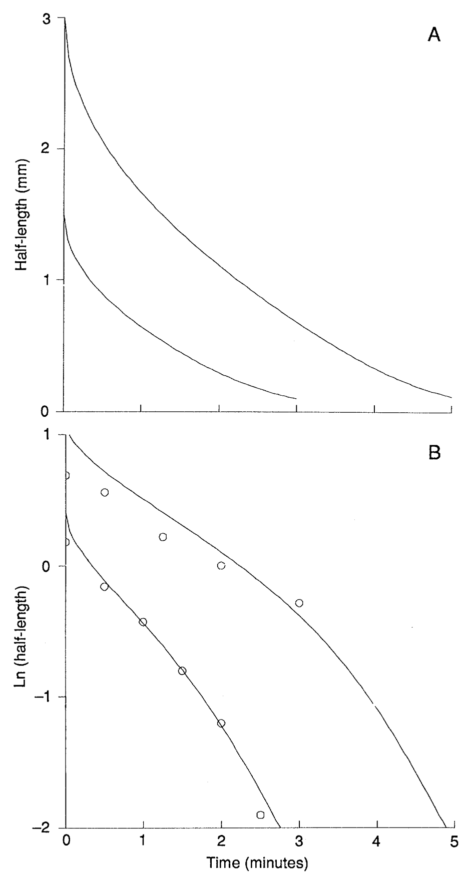

Fig. 3. Life history of an elongated spiral. (**A**) Numerical solution of Eq. 2, with $D = 0.12$ mm^2/min and $d = 1$ mm. (**B**) Same as (A), plotted semilogarithmically. The circles record the approximate length of two spirals in a series of photographs of scroll waves in BZ reagent.

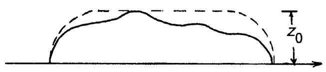

Fig. 4. Filament of an elongated target pattern. Both ends of the filament (solid line) terminate at the same boundary. If the filament obeys $N = D\kappa$, it will collapse in finite time. The lifetime of such an elongated target pattern is bounded above by the lifetime of the comparison curve (dashed line) consisting of a straight line with quarter-circular ends.

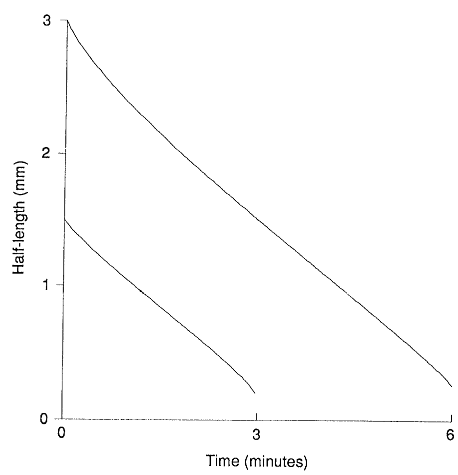

Fig. 5. Half-length of an elongated target pattern as a function of time. The equation of motion was solved numerically, starting with a semielliptical initial filament with maximal height 0.5 mm and initial half-lengths of 1.5 and 3 mm, with $D = 0.12$ mm^2/min.

this curve initially will do so for all time. This is because the only point where the comparison curve moves with velocity equal to D times its curvature is at the end points and along the straight line. Everywhere else its normal velocity is less than D times its curvature. Thus if the end points are initially a distance x_0 apart, the time until collapse of the structure is $T_c < x_0 z_0 / 2D$.

This treatment suggests that elongated target patterns (hot dog sources) should shrink at a rate no slower than the constant rate $2D/z_0$. Numerical simulations confirm this estimate and show further that the collapse is nearly linear in time. Figure 5 shows the half-length as a function of time starting from a semielliptical filament with height 0.5 mm. For this filament the rate of collapse of the length was 0.81 mm/min compared with the estimate of 0.48 mm/min. From Winfree's time-lapse records of BZ waves, we measured the shortening of 11 hot dog sources and found that they did indeed shrink at a roughly constant rate (seven at 1 mm/min, three at 2 mm/min, and one at 0.5 mm/min).

Both hot dog sources and scroll rings have finite lifetimes, but hot dog sources disappear much more quickly than comparably sized scroll rings. A hot dog source with initial length of 5 mm would have a lifetime of ~5 minutes, but a full scroll ring with the same initial diameter 5 mm would have a lifetime of ~26 minutes. Furthermore, the hot dog source shrinks linearly in time (for example, to half its initial length in 2.5 minutes), whereas a full scroll ring shrinks more slowly at first (for example, to half its initial diameter in 20 minutes). Thus the shrinking of hot dog sources should be much more noticeable than the shrinking of full scroll rings.

The equation of motion for an untwisted untorted scroll wave suggests that elongated spirals will evolve nearly exponentially into well-shaped (symmetric) spirals, whereas hot dog sources are unstable and will disappear in finite time. The time courses of these changes as predicted by the theory are in good agreement with experimental observations. The equation of motion $N = D\kappa$, therefore, gives a simple quantitative description of some features of scroll wave evolution. However, this equation of motion is incorrect in several respects. For one, it does not account for "drift" of scroll rings in a direction perpendicular to the plane of the ring (*13, 16, 18*). Furthermore, nonplanar filaments have torsion and twist, but these variables are not accounted for by $N = D\kappa$. Third, it is observed numerically that under certain conditions scroll rings expand instead of shrink (*18*). This behavior is also inconsistent with the equation $N = D\kappa$, and has so far eluded theoretical understanding.

REFERENCES AND NOTES

1. A. T. Winfree, *Science* **175**, 634 (1972).
2. J. D. Gross, M. J. Peacey, D. J. Trevan, *J. Cell Sci.* **22**, 645 (1976); K. J. Tomchik and P. N. Devreotes, *Science* **212**, 443 (1981).
3. M. A. Allesie *et al.*, *Circ. Res.* **333**, 54 (1973); A. T. Winfree, *Sci. Am.* **248**, 144 (May 1983).
4. V. I. Koroleva and J. Bures, *Brain Res.* **173**, 209 (1979); J. Bures, O. Buresova, V. I. Koroleva, in *Neurological Mechanisms of Epilepsy*, V. M. Okujava, Ed. (Metsnierba, Tbilisi, 1980), p. 120.
5. A. T. Winfree, *Science* **181**, 937 (1973).
6. ———, *Faraday Symp. Chem. Soc.* **9**, 38 (1975).
7. B. J. Welsh, J. Gomatam, A. E. Burgess, *Nature (London)* **304**, 611 (1983).
8. A. T. Winfree and S. H. Strogatz, *ibid.* **311**, 611 (1984).
9. A. T. Winfree, *Sci. Am.* **230**, 82 (June 1974).
10. A continuous space curve is described by a "moving trihedral" of three unit vectors, the tangent (**T**), normal (**N**), and binormal (**B**), which are functions of arc length (s) along the curve. By definition, curvature (κ) is $d\mathbf{T}/ds \cdot \mathbf{N}$ and torsion (τ) is $d\mathbf{B}/ds \cdot \mathbf{N}$. The twist $d\phi/ds$ of a scroll wave rotating around such a space curve is the change in phase (ϕ) with respect to arc length measured relative to the axes of the normal-binormal plane.
11. V. S. Zykov, *Biophysics* **25**, 906 (1980); *Modelling of Wave Processes in Excitable Media* (Manchester Univ. Press, Manchester, U.K., 1987).
12. J. P. Keener, *SIAM J. Appl. Math.* **46**, 1039 (1986); ——— and J. J. Tyson, *Physica* **21D**, 307 (1986).
13. A. V. Panfilov, A. N. Rudenko, V. I. Krinskii, *Biofizika* **31**, 850 (1986).
14. J. P. Keener, unpublished results.
15. K. A. Brakke, *Mathematical Notes* (Princeton Univ. Press, Princeton, NJ, 1978), vol. 20.
16. A. V. Panfilov and A. M. Pertsov, *Dokl. Biophys.* **274**, 58 (1984).
17. A. T. Winfree, unpublished photographs.
18. A. V. Panfilov and A. N. Rudenko, *Physica* **28D**, 215 (1987).

19. This work was supported in part by NSF grant DMS-8601134 (J.P.K.) and NSF grant MCS 83-01104 (J.J.T.). We thank A. T. Winfree who kindly made available to us his time-lapse photographs of scroll waves in BZ reagent.

25 September 1987; accepted 11 January 1988

A Cesium-Selective Ion Sieve Made by Topotactic Leaching of Phlogopite Mica

SRIDHAR KOMARNENI* AND RUSTUM ROY

A hydrated, sodium phlogopite mica with a *c*-axis spacing [*d*(001)] of 12.23 Å made by careful low-temperature leaching, showed extremely high selectivity for cesium. This newly discovered cesium ion sieve is useful in the decontamination as well as immobilization of cesium at room temperatuıe through chemical bonding and may find applications in nuclear waste disposal and in decontamination of the environment after accidental releases of nuclear materials.

THE PRINCIPAL LONG-TERM PROBLEM caused by nuclear reactor accidents is the contamination of the environment with radioactive ^{137}Cs, as was evidenced by the Chernobyl nuclear reactor accident (*1*). This is because cesium is very volatile and can be carried long distances. Decontamination of the environment, humans, and animals is possible with the use of a highly selective cesium ion sieve either by dispersion, for example, in water or soil, or by ingestion by humans and animals.

Zeolites and clay minerals are naturally occurring cation exchangers that have been

Materials Research Laboratory, Pennsylvania State University, University Park, PA 16802.

*To whom correspondence should be addressed.