

# Chemical Waves

## Diffusion + Reaction Kinetics



## Chemical Waves

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**Abstract:** In our paper we try to describe the basic concepts of chemical waves and spatial pattern formation in a simple way. We pay particular attention to self-organisation phenomena in extended excitable systems. These result in the appearance of travelling waves, spiral waves, target patterns, Turing structures or more complicated structures called scroll waves, which are three-dimensional systems. We describe the most famous oscillating reaction, the Belousov–Zhabotinsky (BZ) reaction, in greater detail. This is because it is of great interest in both physical chemistry and in studies on the evolution and sustenance of self-organising biological systems.

**Keywords:** Belousov–Zhabotinsky reaction • chemical waves • oscillating reactions • pattern formation • Turing structures

### Introduction

This is a work about the waves of chemical reactions that can travel through the reaction medium, and the important stationary patterns that may spontaneously form in some particular chemical systems.<sup>[1]</sup> We concentrate on oscillating reactions<sup>[2]</sup> and the so-called “reaction–diffusion systems”. Spatial organisation phenomena occur when a chemical reaction, with an autocatalytic step (or feedback–retroaction loop) in its reaction mechanism, is coupled with the diffusion of species. In reaction–diffusion systems molecules react together when they collide, and, as a result of the var-

iation in the concentrations of the components, chemical waves propagate. This class of chemical system has been extensively studied, as it has a wide variety of biological implications. Some systems are able to create a stationary pattern of stripes with high chemical concentration alternating with stripes of low concentration. Many scientists think that the natural models from which these systems are formed have much to teach us about the spatial schemes that occur in the development of plants and animals. For example, alternating stripes of different colours that develop in oscillating chemical systems can help us to understand many aspects of the morphology of both simple and complex organisms.

### Target Patterns in Excitable Media

Propagating waves may develop in excitable media. These are spatially distributed systems that have the ability to propagate signals without damping, and that cannot support the passing of another wave until a certain amount of time has passed (this is known as the refractory time). An example of an excitable medium is a forest. If a wildfire burns through the forest, it travels as a wave from its initiation point, and regenerates with every tree it ignites and thus no fire can return to a burnt spot until the vegetation has gone through its refractory period and regrown. By contrast, passive wave propagation is usually characterised by a gradual damping of signal amplitude due to friction, as happens, for example, to sound waves passing through air.

Excitable media are widespread in physics, chemistry and biology. Among the most important examples of active media are the autocatalytic chemical reactions,<sup>[3]</sup> the most well studied being the Belousov–Zhabotinsky (BZ) reaction.<sup>[4–7]</sup> Coloured patterns<sup>[8–11]</sup> can arise when these types of reaction are carried out in thin layers, such as in a Petri dish. They correspond to spatial variations in chemical concentrations. In its classical form the BZ reaction consists of a one-electron metal redox catalyst (usually  $\text{Ce}^{3+}/\text{Ce}^{4+}$  or  $\text{Fe}^{2+}/\text{Fe}^{3+}$ ), an organic substrate (usually malonic acid,  $\text{HOOC-CH}_2\text{-COOH}$ , or citric acid) that can be easily brominated and oxidised, and bromate ions. All of these are dis-

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solved in sulphuric or nitric acid.<sup>[12,13]</sup> What we observe in experiments is that if we pour a thin coat (of about 1 mm) of the BZ solution gently into a Petri dish, blue concentric circles form and develop (Figure 1). These indicate local re-

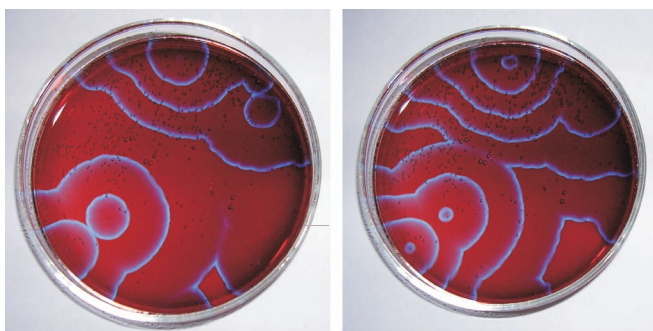


Figure 1. Concentric circular wave fronts for the BZ system catalysed by ferroin in a Petri dish. These two snapshots are taken in succession; there is a three minute time interval between them.

gions of high  $\text{Fe}^{3+}$  concentration that travel through a red background of  $\text{Fe}^{2+}$ , forming target patterns. Generally, a wave of oxidation starts from a leading centre and propagates outwards through the reduced medium in the form of a circular wave. After a certain time  $t$ , a second wave originates at the leading centre and follows the first, making a target pattern propagating radially from the centre. In about 15 min the phenomenon occupies the whole space.

The speed of propagation of the waves is usually roughly constant. When two waves collide they annihilate one another. Target patterns<sup>[4,14]</sup> are also called “travelling waves”, because they are a phenomenon that involves the propagation of concentration fronts and particular chemical waves moving in time.

### Travelling Waves

In general, a wave is characterised by the shape and speed of propagation of the front. This can usually change continually, without any restrictions. By contrast a “travelling wave” is normally defined as a wave that travels without change of shape. To elucidate, let us consider a simple one-dimensional scalar case. If a function  $u(x,t)$  represents a travelling wave, the shape of the function will be the same for the whole time and the speed of propagation of this shape is a constant, which we can call  $c$ . Looking at this wave in a travelling frame moving at speed  $c$ , it will appear stationary. Thus, an observer moving at the same speed in the direction of motion of the wave sees an unchanging picture, which he might describe alternatively as  $u(z)$ .

To be more precise, if the function is that given in Equation (1), then  $u(x,t)$  is a travelling wave, and it moves at constant speed  $c$  in the positive  $x$  direction. Clearly, if  $x-ct$  is constant then so is  $u$ , and this means that the coordinate system moves with speed  $c$ .

$$u(x,t) = u(x-ct) = u(z) \quad (1)$$

$$z = x-ct$$

Thus, travelling wave fronts of chemical reactions are characterised by uniform speed and a constant concentration profile. We will also consider “pulses” and periodic wave trains (see Figure 2). Hence we will discuss three different types of waves.

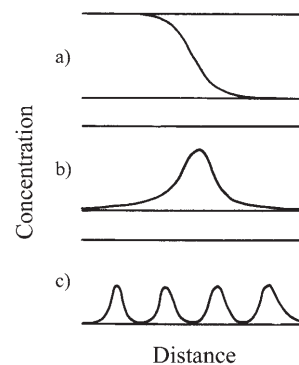


Figure 2. Different types of travelling waves: a) a wave front, b) a pulse and c) a periodic wave train.

- 1) A “front” usually converts the reactants in products so that the compositions ahead of and behind the wave are quite different (Figure 2a). It acts as a flame does.
- 2) We speak of a “pulse”, when an intermediate is firstly produced by a certain front and then it is converted back by a second recovery wave (Figure 2b). In this way, the intermediate concentration is not the same ahead of and behind the pulse.
- 3) Finally, the combination of a series of fronts or pulses that are continuously being initiated at some point or “centre” and which follow each other in the medium are referred to as “periodic wave trains” (Figure 2c).

Though the systematic study of macroscopic chemical waves and patterns originating from the coupling of reaction kinetics and diffusion is quite recent, the first studies in this field were initiated by Robert Luther<sup>[15]</sup> one century ago. In 1906 he wrote an important paper on propagating fronts in autocatalytic back-reactions. His paper is the first clear report of moving waves of reactions being possible in homogeneous liquid-phase chemical systems. Some years later, Anatol Zhabotinsky<sup>[6]</sup> contributed greatly to the growth of this new discipline when he noticed that in thin layers of the oscillating solution, leading centres of oxidation arise spontaneously and organise the two-dimensional medium into target patterns. Today pattern formation is one of the subjects of most interest to chemists and physicists, and is frequently studied.

Of course spontaneous pattern formation is not restricted to chemical systems. For example, it is also common in dif-

ferent biological systems. In biology the appearance of travelling waves of chemical concentration seems to be a key element or precursor to a developmental process in a vast number of phenomena. Propagating wave forms of varying biochemical concentrations are one means for continual interchange of information at both inter- and intracellular level.

### Spiral Waves

Initial conditions in the excitable media can give rise to a different pattern of excitation called a spiral wave.<sup>[16,17]</sup> These types of wave appear in a vast range of chemical, biological and physical systems, which can be considered as effectively two-dimensional excitable or oscillatory media in which the local nonlinear dynamics exhibit threshold behaviour, separating an excited state from a recovered state. The waves of excitation spread in a diffusive manner, starting from one region and triggering the neighbouring one; they do not decrease as they travel through the stationary medium. Thus a single spiral wave in a large homogeneous medium acts as an organising centre, imposing its pattern throughout the medium. What happens exactly is that spiral waves form naturally when a wave front travels around a pivot point, repeatedly re-exciting itself. A spiral is created as points further from the pivot have to travel farther to make a circuit, and therefore lag behind the wave at the centre. By contrast the pivot point around which the spiral rotates, generally defined as the central core, is not invaded by the propagating wave.

The mechanisms of the onset and stability of spirals are the subjects of extensive ongoing investigation.

The free end of the spiral is also called the “spiral tip”. Its shape, together with the excitability of the media, influences the propagation speed near the spiral tip and, consequently, the whole geometry and period of the spiral waves. The tip motion of a rigidly rotating spiral propagating in an isotropic medium can be complex, meandering quasi-periodically within a bound region, or hyper-meandering in an unbound region; any inhomogeneities and/or boundary interactions can produce drift.<sup>[18,19]</sup> The activity along any radii far from the core enclosed by the tip spiral is seen as a periodic wave train with a temporal frequency  $\omega$ , corresponding to the rotation frequency of the spiral, and spatial wavelength  $\lambda$ . Generally,  $\omega$  and  $\lambda$  are single-valued functions of the medium parameters. The wave front approximates to an involution of a circle with a pitch  $\lambda$ , which in the simplest case, that is, a one-armed spiral, is a periodic function of phase  $\varphi$ . In terms of polar coordinates  $(\theta, r)$  the phase can be written as Equation (2), with  $a > 0$  giving the wavelength  $\lambda$  and pitch  $dr/d\theta|_{\varphi=\text{const}}$  equal to  $1/a$ .

$$\varphi = \omega t \pm \theta + a(r - r_0) \quad (2)$$

When subjected to parametric perturbation, a rotating spiral can undergo destabilising changes that annihilate the

wave. The form of the spiral can be restored, but its location and phase may be changed. One of the most important and most studied types of destabilising change is the spontaneous wave breaking on the spiral arm. This effect is often called “spiral break up”; it does not annihilate the activity, but shows itself rather in multiple cores of spirals that propagate in an erratic way through the medium.

The process by which a wave front breaks up is not yet clear, and it is still under investigation.

The annihilation of spirals could be due to the meandering of the wave tip in homogeneous media, which may cause the wave to collide with a boundary, and to spirals that may drift to the boundary in heterogeneous media.

The three-dimensional structures typically generated in these media have also been studied. These are called “scroll waves”.<sup>[20,21]</sup> They may become unstable in weakly excitable media, giving rise to a turbulent regime. Cardiac fibrillation may be an example of this. As stated above, the existence and formation of spiral waves of excitation (sometimes also called “vortices” or “rotors”) is a general property of excitable media of different natures. In some applications the propagation of waves in excitable media is not always seen as desirable. For example, cardiac tissue<sup>[22–24]</sup> is also considered an active media. The spontaneous break up of a spiral wave into several waves and their subsequent multiplication can lead to spatiotemporal chaos. Such dynamics have been thought to be at the base of the mechanism for the onset of ventricular fibrillation in the heart and sudden cardiac death. Thus developing valid methods for suppressing travelling waves in excitable media is of very great importance.

Spirals and interacting spirals can be initiated in a thin-layer BZ solution in a Petri dish. Observations of inwardly rotating spirals found in different Belousov–Zhabotinsky systems have also been reported.<sup>[25,26]</sup> These last types of spiral waves are also called “antispirals”. An example is shown in Figure 3.

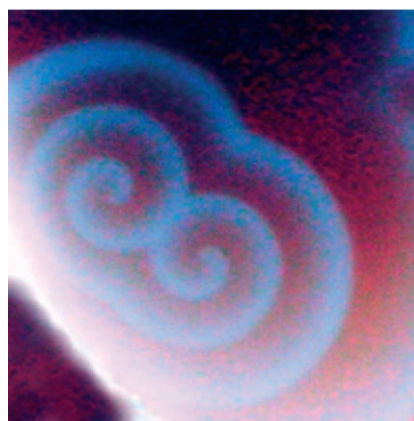


Figure 3. Inwardly rotating spiral waves occur when a  $\text{Fe}^{\text{II}}/\text{Fe}^{\text{III}}$ -catalysed BZ system is performed in the presence of lipids. To be precise, the picture shows an antispiral which appeared at the edge of the Petri capsule in a BZ/DPPC (DPPC = 1,2-dipalmitoyl-*sn*-glycero-3-phosphocholine) system when the lipid content was 18% w/w.

Spiral patterns have also been observed in other very important systems, such as aggregating slime molds,<sup>[27]</sup> developing oocytes,<sup>[28]</sup> spreading depression in chicken retina,<sup>[29]</sup> Rayleigh–Bénard convection,<sup>[30]</sup> catalytic crystal surfaces<sup>[31]</sup> and galaxies.<sup>[32]</sup>

### Turing Structures

We have just seen that coloured spatial patterns, which correspond to spatial variations in chemical concentrations, may arise when a reaction is carried out in thin layers. These patterns move with time. There are, however, other important types of patterns called “Turing structures”, which are stationary in time and periodic in space, or periodic in both time and space. Turing structures are beautiful patterns and they too are very common in biological and chemical systems.

The name of these structures comes from an English mathematician, Alan Turing, who studied them in depth. In 1952, he wrote an important paper<sup>[33]</sup> that provided the basis of the theory of morphogenesis (morphogenesis is the development of pattern and form in living systems). He suggested that chemicals (which he also called “morphogens”) can react and diffuse in such a way as to produce a spatial pattern in morphogen concentration. Indeed the stationary concentration patterns only originate through coupling of reaction and diffusion processes.

Turing realised that diffusion is not always a homogenising influence which avoids chemical gradients and leads to uniform spatial distributions, as we instinctively assume from daily experience. For example, if we put a drop of ink in a beaker full of water, we see that the ink eventually invades all the solution. A spontaneous transfer takes place from the zones of high concentration to those of lower concentration, but the situation is completely different when we consider reaction–diffusion systems, such as self-catalytic reactions.

Thus, it was some surprise to discover that diffusion can have a contrary effect, producing chemical gradients, and that spatial patterns can be formed by reaction–diffusion systems by combining local activation with long-range inhibition. This concept was revolutionary at that time, but crucially important.<sup>[34]</sup>

The key elements, the suitable combination of which could give rise to chemical pattern formation, are:

- 1) Two or more chemical species.
- 2) Different diffusion rates of the participants.
- 3) Chemical interactions.

The kinetics should include a positive feedback, such as autocatalysis, on a species called an “activator” and an inhibitory process. The inhibitor’s diffusion coefficient should be much greater than that of the activator. Reactions that show bistable or oscillatory behaviour usually fulfil these conditions.

Turing structures in reaction–diffusion systems were first investigated theoretically and then modelled by numerical simulations.<sup>[35]</sup>

The basic equation for Turing reaction–diffusion mechanisms is given in Equation (3) in which  $\mathbf{c}$  is the vector of morphogen concentrations,  $f$  represents the reaction kinetics and  $D$  is the diagonal matrix of positive constant diffusion coefficients.

$$\partial \mathbf{c} / \partial t = f(\mathbf{c}) + D \nabla^2 \mathbf{c} \quad (3)$$

In any practical case the kinetics are always nonlinear. In the absence of diffusion and of all the phenomena of convection,  $\mathbf{c}$  tends to a linearly stable uniform steady-state, while in certain conditions spatially inhomogeneous patterns can evolve as a result of diffusion driven instability, if the diffusion coefficients are very different.

### Examples of Turing Structures and Waves in Real World Systems

Turing structures and their dynamics have been widely studied in a huge variety of systems. We describe here two particular chemical systems in which waves and stationary patterns can spontaneously appear.

The most widely studied reaction–diffusion chemical system is the chlorite/iodide/malonic acid (CIMA) reaction and its numerous modified versions. In these Turing structures evolve spontaneously.<sup>[36]</sup> Indeed in the CIMA reaction, which is sometimes performed in gel with starch as an indicator, some striped and hexagonal (spotted) structures (see Figure 4)<sup>[37]</sup> were observed in the reactor. This is because

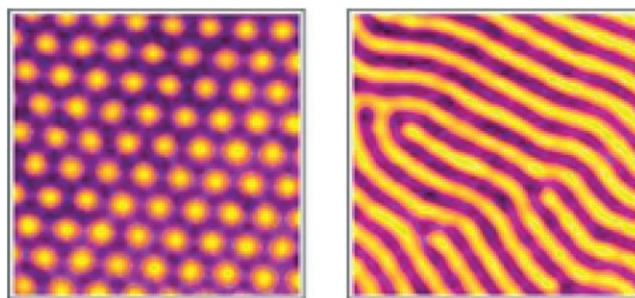


Figure 4. Examples of Turing structures: honeycomb (left) and labyrinthine (right) structures. (The photographs are reproduced with the permission of P. De Kepper and co-workers, who work at Centre de Recherche Paul Pascal, in Bordeaux, and are taken from their Web site: <http://www.crrpp.u-bordeaux.fr/>.)

the starch forms a complex with iodide, and iodine (the activator species) and the starch–triiodide complex diffuse much more slowly in the gel than the chlorite or chlorine dioxide, which acts as the inhibitor species.

Another important study on the spatially extended oscillatory BZ reaction and its importance for the comparison of the evolution and sustenance of self-organising biological

systems started at the end of the eighties. It was based on the incorporation of the standard Belousov–Zhabotinsky chemical oscillator into a range of hydrotropic,<sup>[38,39]</sup> micellar<sup>[40,41]</sup> and reverse micellar<sup>[42]</sup> systems under batch conditions. The reverse micelles of amphiphiles in organic media (first studied by Balasubramanian and Rodley,<sup>[43]</sup> and then by Gonda<sup>[44]</sup>) contain an internal water pool of defined size, shape and solvent properties. The ferroin-catalysed Belousov–Zhabotinsky reaction system in water-in-oil microemulsions with anionic surfactants, for example, aerosol diisooctylsulfosuccinate (AOT; that is, reverse micelles of sodium diisooctylsulfosuccinate), has been studied in great detail by Epstein, Vanag and co-workers.<sup>[25,45–47]</sup> The AOT surfactants do not react chemically with any of the BZ reagents, and they are not brominated or destroyed during the reaction. Chemical oscillations are found in the BZ–AOT system in the pseudo aqueous phase, even though its pH is extremely high in this stage. It has been found that manifestation of the BZ oscillations in the AOT micelles depends on the size and concentration of microemulsion droplets. However Turing structures are not found only in spherical droplets.

Recently new versions of the Belousov–Zhabotinsky reaction–diffusion system carried out in layered water domains showed particularly interesting spatial structures, such as in BZ–AOT microemulsions. For example, a BZ reaction carried out in anisotropic environment of 1,2-dipalmitoyl-*sn*-glycero-3-phosphocholine (DPPC)/water binary system showed specific patterns, such as striped standing waves, inwardly rotating spirals and the same Turing structures.<sup>[26]</sup>

The appearance of the different structures depends on the DPPC concentration (see Figure 5).

It was found that if the DPPC concentration was kept between 7 and 10% w/w, then travelling waves and pacemaker structures were obtained; many other interesting patterns were found when the DPPC concentration increased. Turing structures appeared when the DPPC concentration was below 30% w/w. At greater concentrations all systems exhibited only a few blue-filled circular spots, and no waves or other interesting structures were observed.

The examples of Turing structures in nature are also very numerous, and they have already been the subjects of interesting studies. Here we are referring to spots and stripes on animal coats (see Figure 6),<sup>[48,49]</sup> patterning on sea shells,<sup>[50]</sup> stripes on tropical fish,<sup>[51]</sup> alligator teeth,<sup>[52]</sup> the labyrinthine



Figure 6. Turing structures in the coats of leopards, cheetahs and zebras, and stripes on tropical fish.

patterns of the cerebral cortex of mammals<sup>[53]</sup> (see Figure 7) and many other examples. In this case too, Turing instability, which is based on activation and inhibition kinetics of the diffusing chemical species, has been proposed as the basic mechanism through which, for example, stripes on the tiger or the patterns on sea-shells form.

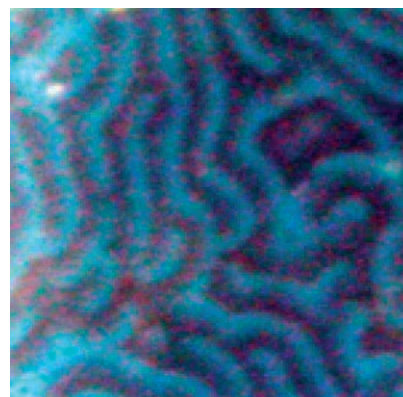


Figure 7. Labyrinthine patterns, which look like a handful of squirming worms, in a BZ reaction–diffusion system. They are similar to the cerebral cortex of a human brain.



Figure 5. From left to right: travelling waves, labyrinthine structures, inwardly rotating spirals, striped standing waves, and blue-filled circular spots. These were observed as the lipid concentration increased in a BZ reaction carried out in anisotropic environment of 1,2-dipalmitoyl-*sn*-glycero-3-phosphocholine (DPPC)/water binary system.

## Conclusion

Firstly we have attempted to show, in a general way, the major characteristics of excitable media, such as oscillating chemical reactions, and some important concepts necessary for understanding their behaviour. The most interesting property of excitable media is the existence of autowaves, that is, nonlinear spatiotemporal structures propagating through the medium. In chemical systems, autowave patterns are one of the most evident examples of self-organisation in space and time.

We introduced the Belousov–Zhabotinsky reaction as an excellent example of an oscillating reaction. The BZ reaction's capacity of spontaneous spatiotemporal auto-organisation is described. This gives rise to the formation of chemical (concentric and spiral) waves and interesting stationary patterns (Turing structures). The BZ reaction has been studied for many years and is well understood; consequently it is considered the prototype oscillator, which can also help us to understand oscillations in the biological field. Pattern formation in reaction–diffusion systems (such as the BZ reaction) have, indeed, for some time been considered the mechanism for morphogenesis. Spiral waves of the BZ reaction have also been observed in other types of media, such as intact and cultured cardiac tissue, mold aggregates and so forth. Alan Turing suggested that while diffusion alone tends to create uniform states, when it is coupled with chemical reactions, spatial patterns in chemical composition may appear. This mechanism has also been suggested for the formation of natural patterns, such as animal stripes or the regular shape and rearrangement of leaves.

Investigation of these oscillating reactions, a self-organisation in space and time, and reaction–diffusion systems is very important; firstly because they can be used as models for helping us to understand more complex systems and secondly because they can help to validate the theoretical concept of irreversible process far from the equilibrium.

Indeed, although the BZ reaction is a chemical rather than biological oscillator, understanding its mechanics will also help us to understand biological oscillations such as the beating of the heart.<sup>[22–24]</sup> The dynamics of spiral waves, for example, is thought to be related to the onset of ventricular fibrillation. It seems that the precursor to fibrillation is the appearance of rotating waves of electrical impulses.

Finally, we report further observations of spiral and target waves and Turing patterns in the Belousov–Zhabotinsky reaction dispersed in water nanodroplets of a water-in-oil microemulsion, and in BZ/DPPC systems. The latter are the first model for a biological structure. The last study in particular, and the possibility that peculiar patterns can also form in layered water domains, should stimulate further research on these types of system, which increasingly mimic analogous phenomena in vivo in biological systems.

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