

Turing Patterns in the Chlorine Dioxide-Iodine-Malonic Acid Reaction–Diffusion Batch System

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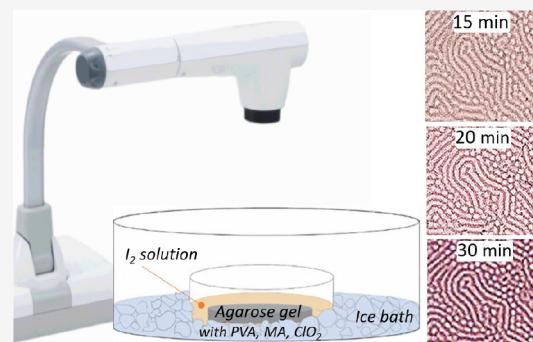
ABSTRACT: Under the appropriate conditions, oscillatory chemical reactions have the capacity to generate chemical waves and spatial patterns. Among these structures, Turing patterns are a distinct class that, to date, has not been commonly demonstrated in a classroom environment. We present here a novel, practical procedure for the demonstration of Turing patterns in a chemical reaction–diffusion batch system, which has the potential for diverse applications in a variety of settings, while allowing for a degree of variability. Altering the experimental conditions can produce varying proportions of spotted and striped patterns that are stable for long periods of time, allowing them to be viewed for entire lecture periods. This remarkable demonstration can be a valuable pedagogical tool for the introduction of a variety of chemical concepts, including reaction–diffusion kinetics, nonequilibrium thermodynamics, and autocatalysis.

KEYWORDS: Undergraduate/Gallery Education, Physical Chemistry, Kinetics, Reaction–Diffusion, Chemical Patterns

INTRODUCTION

Nonlinear chemical dynamics comprises the study of chemically reacting systems that display complex dynamical phenomena, such as temporal oscillation, chaos, and spatiotemporal pattern formation.¹ Its consolidation as a field of study in chemistry occurred in the early 1970s with the elucidation of the Belousov–Zhabotinsky (BZ) reaction mechanism and the spread of new concepts of nonequilibrium thermodynamics developed in the 1950s.^{2–6} In general, these works were fundamental in proving that chemical oscillations could emerge through uni- and bimolecular reaction steps and that the appearance of this class of phenomena is consistent with physical law.⁷ Moreover, they also identified two necessary conditions for a chemical reaction to exhibit complex behavior: 1) the system should be kept far from equilibrium and 2) it must contain positive feedback, usually autocatalysis, and negative feedback.^{6,8} The comprehension of these conditions, in addition to the concepts of nonlinear dynamics theory, helped scientists discover a number of new chemical oscillators.^{1,6,9} The Briggs–Rauscher (BR) and BZ reactions are chemical reaction prototypes for a large number of dynamical phenomena, including temporal oscillation, chaos, and waves.¹⁰ Due to their wide variety of behaviors, color changes that create alluring patterns, and relatively simple experimental setup, the BR and BZ reactions are widely employed in lecture demonstrations.

Classroom demonstrations of oscillatory chemical reactions have become a valuable pedagogical tool to introduce students to a variety of chemical concepts including kinetics,



thermodynamics and catalysis.¹¹ Over the past 50 years, nonlinear chemical dynamics has been the subject of numerous articles in this *Journal*.^{11–41} Such works span experimental demonstrations of oscillations and waves through the BZ and BR reactions,^{12,15,16,18,21,31} the mercury beating heart,³⁸ the gas evolution oscillator,¹⁷ the chemiluminescent oscillator,^{11,20} pH oscillators,¹⁴ the salt-water oscillator,^{39,40} the explosion oscillator,²² and the Liesegang rings,^{42–45} to the theoretical presentation of the foundations of nonlinear dynamics,^{13,23,25,28,37} and nonequilibrium thermodynamics.¹² Most of these classroom demonstrations focused on oscillations in stirred systems and propagating waves in unstirred systems. However, there are other structures that can emerge from similar dynamic systems and are closely related to the emergence of life. Examples of such structures are Turing patterns, which are stationary in time and periodic in space.^{1,6}

Alan M. Turing, a British mathematician, predicted the occurrence of stationary patterns in chemical systems in his 1952 groundbreaking work “The chemical basis of morphogenesis.”⁴⁶ Using a hypothetical chemical model of a reaction–diffusion system, Turing determined that the system could have a spatially uniform steady state that is simultaneously

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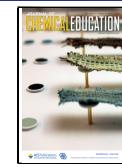


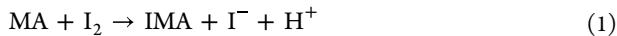
Table 1. Preparation of Materials

	Reagent	Amount	Final Concentration	Total Volume
Solution A	Polyvinyl Alcohol (PVA) (CAS 9002-89-5)	1.0g	4% (by weight) PVA	25 mL
	malonic acid (MA) (CAS 141-82-2)	0.0104 g	4 mM MA	
Solution B	1 M sulfuric acid (H_2SO_4)	0.5 mL	20 mM H_2SO_4	25 mL
	chlorine dioxide (ClO_2) ^a	0.5 mL	4 mM ClO_2	
Solution C	1 M H_2SO_4	0.0207 g	20 mM H_2SO_4	25 mL
	iodine (I_2) (CAS 7553-56-2)	2.5 mL	3.2 mM I_2	
	glacial acetic acid	0.5 mL	N/A	
Agarose gel ^b	agarose (CAS 9012-36-6)	0.3 g	2% (by weight) agarose	N/A

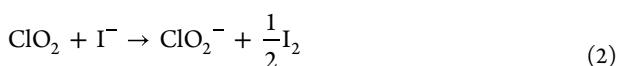
^aThe amount of ClO_2 used to prepare solution B varies depending on the concentration of the stock solution. A UV-vis spectrophotometer can be used to determine the ClO_2 concentration of the stock solution (ClO_2 molar absorptivity = $1200\text{ M}^{-1}\text{cm}^{-1}$ at 360 nm). If only a VIS spectrophotometer is available, the molarity of the ClO_2 stock solution can be estimated at 390 nm. At this wavelength, the molar absorptivity of ClO_2 is $557\text{ M}^{-1}\text{cm}^{-1}$. ^bA gel disc with the desired thickness of 3.2 mm is prepared by using rubber O-rings as molds placed between two glass plates. Weights are placed on the top glass during the gel solidification to maintain the gel thickness. After the gel has solidified, the gel disc is cut using a 1–5/8" arch punch. A 4 cm diameter of the gel disc is recommended, but slightly larger or smaller gel diameters can also be used.

stable and unstable to homogeneous and nonhomogeneous perturbations, respectively.^{6,8,46} In his model, local fluctuations would grow to form a stationary, patterned state resulting in a symmetry-breaking phenomenon.⁴⁷ Turing observed that the emergence of these structures requires a mechanism composed of an activator, i.e., a chemical that increases the rate of its own production, and an inhibitor, i.e., a chemical that decreases the rate of the activation process, typically by removing an autocatalytic species. Moreover, in such a reaction–diffusion process, the inhibitor must diffuse significantly faster than the activator.^{6,8,46} This last condition is difficult to fulfill experimentally because in aqueous solution small molecules and ions share similar diffusion coefficients on the order of $10^{-5}\text{ cm}^2\text{s}^{-1}$. It took almost 40 years after Turing's work for chemists to overcome this issue and to observe Turing patterns in the chlorite-iodide-malonic acid (CIMA) reaction.^{6,48}

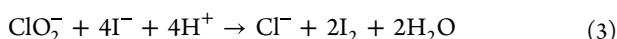
A modified version of the CIMA reaction, the chlorine dioxide-iodine-malonic acid (CDIMA) reaction, is widely employed in the construction of reaction–diffusion systems for studying Turing patterns. The chemistry of the CDIMA reaction can be described by three overall processes with the corresponding rate laws:^{6,49}



$$r_1 = \frac{k_{1a}[\text{MA}][\text{I}_2]}{k_{1b} + [\text{I}_2]}$$



$$r_2 = k_2[\text{ClO}_2^-][\text{I}^-]$$



$$r_3 = k_{3a}[\text{ClO}_2^-][\text{I}^-][\text{H}^+] + \frac{k_{3b}[\text{ClO}_2^-][\text{I}^-][\text{I}_2]}{\alpha + [\text{I}]^2}$$

where MA is malonic acid and IMA is iodomalonic acid, i.e., $\text{CHI}(\text{COOH})_2$. Process 1 summarizes the reaction between malonic acid and iodine to produce iodide, which is the activator of the reaction.⁴⁹ Process 2 represents the reduction of chlorine dioxide by iodide, producing chlorite ions, which play the role of the inhibitor required for Turing instability by removing the activator in the next step.⁴⁹ Process 3 comprises a feedback process that is crucial for the dynamics of the

CDIMA reaction.^{6,49,50} The rate r_3 includes parameter α , the threshold concentration of $[\text{I}^-]$ beyond which the suppressive impact of iodine becomes significant.⁵¹ Notably, in this reaction step, iodide functions as a self-inhibitor. An elevated iodide concentration inhibits its destruction and indirectly stimulates its own production.⁴⁹

Even though the activation–inhibition mechanism of the CDIMA reaction suggests it can exhibit complex spatiotemporal behavior, in principle it can not generate Turing patterns because the diffusion rates of the I^- and ClO_2^- are quite similar. However, the addition of PVA to the reaction medium decreases the effective iodide diffusivity. The PVA molecules are large and have a very small diffusion coefficient. When they are present in the CDIMA system, iodide ions reversibly bind to PVA in the form of the triiodide-PVA complex PVAI_3^- , which in higher concentrations has a distinct dark purple color:



When iodide is bound to PVA, its effective diffusivity decreases and it diffuses much slower than chlorite. Thus, PVA acts not only as a color indicator for the patterns, but also as a species that allows Turing patterns to form in the CDIMA reaction.^{6,50}

The investigation of Turing patterns is often carried out in open systems using a continuously fed unstirred reactor (CFUR), keeping the system far from equilibrium.⁷ However, it also has been demonstrated that it is feasible to observe transient Turing patterns in the CDIMA reaction in a closed, batch system.^{52–54} The difficulty of a classroom demonstration of such structures lies in the temperature dependence of the CDIMA system, which has to be maintained below 8 °C for Turing patterns to form.^{1,53} Thus, until now, no classroom demonstrations of Turing patterns have been proposed or utilized. This article presents for the first time a workable procedure for producing Turing patterns in a closed system by utilizing the CDIMA reaction with common laboratory equipment in a classroom setting.

MATERIALS AND METHODS

Analytical reagent poly(vinyl alcohol) ((C_2H_4O)_n, PVA, MW 9000–10000), sulfuric acid, malonic acid ($\text{CH}_2(\text{COOH})_2$, MA), and iodine (Sigma-Aldrich) are used without purification, and solutions are prepared with deionized water. The chlorine dioxide stock solution is synthesized from analytical

reagent potassium chlorate ($KClO_3$), oxalic acid ($H_2C_2O_4 \cdot 2H_2O$), and sulfuric acid (Sigma-Aldrich).⁵⁵ The Supporting Information includes the details of the ClO_2 synthesis. The solutions are prepared as described in Table 1.

- Soaking of the 2% agarose gel with reagents:** 8 mL of Solution A is mixed with 8 mL of solution B and the mixture is poured into a 60×15 mm Petri dish with the 2% agarose gel disc. The covered dish should be wrapped in parafilm to limit ClO_2 escape and placed in a refrigerator for at least several hours.

All solutions and gels are stored in a refrigerator. The iodine solution should be protected from light to limit its decomposition.

PROCEDURE

The experimental setup entails an ice bath, a smaller (e.g., 58×15 mm) Pyrex Petri dish, a medium size (e.g., 125×65 mm) crystallizing dish, and the use of a document camera with a screen projector and/or a light table. A temperature of approximately $4^\circ C$ is achieved by immersing the Petri dish with the gel in the ice bath inside the crystallizing dish, see Figure 1. The experiment is initiated by transferring 8 mL of

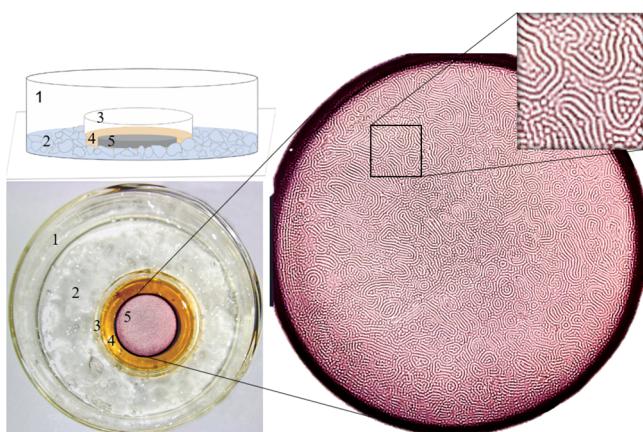


Figure 1. Scheme of the side view and photo of the top view of the demonstration setup and Turing pattern formed in the agarose gel disc with a 40 mm diameter. 1 - Crystallizing dish, 2 - Ice water bath, 3 - Small Petri dish, 4 - Iodine solution, and 5 - Agarose gel disc.

solution C into the Petri dish. The PVA/MA/ ClO_2 -saturated gel is then placed carefully into the Petri dish, followed by the addition of another 2 mL of solution C onto the gel. The final level of the iodine solution is approximately 2 mm above the top of the gel. This positioning ensures a consistent supply of iodine to the gel matrix and contributes to the pattern formation process. If a light table is available, it can be used to enhance the pattern contrast. However, a light source from a document camera, shining from above, is enough to make projected Turing patterns on a classroom screen clearly visible.

HAZARDS

It is recommended to wear appropriate protective equipment, such as gloves and goggles, during the preparation of solutions and demonstrations. Malonic acid has the potential to cause eye damage upon direct contact, and glacial acetic acid is both flammable and capable of causing skin burns upon direct skin contact. Iodine, both a skin and eye irritant and a carcinogen, should not be allowed to come into direct contact with skin due to its staining properties and harmful effects, and a mask should be worn while measuring the iodine. Sulfuric acid, a highly corrosive substance, can severely irritate and burn the skin and eyes and may lead to blindness. Particular care is necessary when preparing stock solutions and dilutions of chlorine dioxide, which is toxic and explosive. Proper disposal of waste liquids should be conducted in accordance with the protocols in the Supporting Information.

RESULTS AND DISCUSSION

The Experiment

Under specific conditions, Turing patterns can arise in a closed batch system suitable for a classroom demonstration. Clear and distinct Turing patterns form when using a 3.2 mm thick 2% agarose gel. The gel, presaturated with 2% PVA, $[MA] = 2$ mM, $[ClO_2] = 2$ mM, $[H_2SO_4] = 20$ mM, is put in a cooled solution of $[I_2] = 3.2$ mM in a Petri dish. The dish is placed in an ice–water bath, as shown in Figure 1. This design ensures that a) no chemical reaction occurs before the beginning of the experiment, b) the gel is loaded with enough chemicals to keep the reaction far from equilibrium for as long as possible, c) there is a sufficient surface area for pattern formation, d) the rates of reactions 1 to 3 are decreased, and e) iodine diffuses

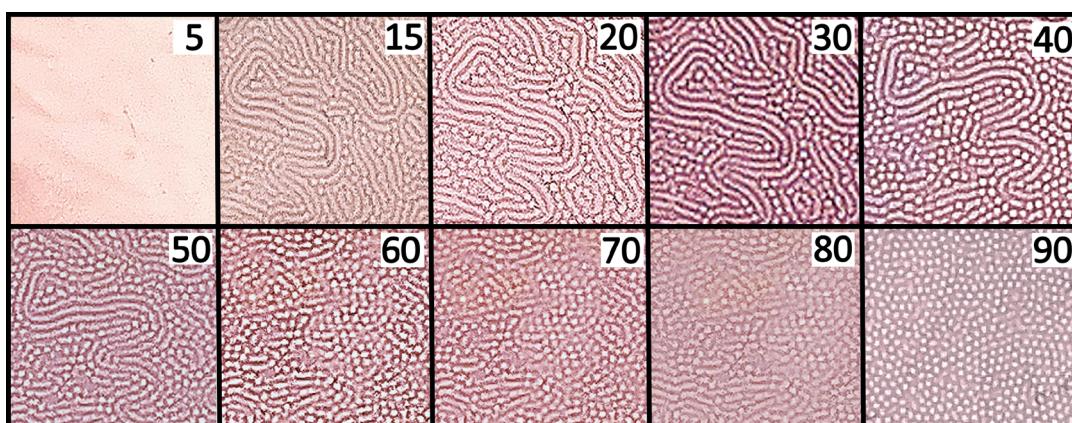


Figure 2. Evolution of transient Turing patterns in a batch system with gel. $[MA] = 2$ mM, $[ClO_2] = 2$ mM. Times (in minutes) are displayed in snapshots, the system size shown is 6×6 mm.

from the solution into the gel, initiating the chemical reactions and forming Turing patterns inside the gel.

Different pattern morphologies are observed in the CDIMA system for different concentrations of reactants. We varied the concentration of ClO_2 in the gels from 1 mM to 2 mM, while keeping the PVA concentration constant at 2% (by weight) and $[\text{I}_2]$ constant at 3.2 mM, and using either 1 mM or 2 mM MA. The patterns formed using 1 mM ClO_2 and 1 mM MA for soaking the gel were primarily composed of spots, and these patterns were stable for only 5 min before fading. A ClO_2 concentration of 2.0 mM with 2 mM MA initially produced predominantly striped patterns, which eventually transitioned into spots. The wavelength of the observed Turing pattern is about 0.4 mm. Consequently, in a classroom demonstration, it is often necessary to enhance the pattern spatial organization and periodicity with a screen projector.

Our findings indicate that a higher concentration of ClO_2 favors stripe pattern formation. Figure 1 depicts a pattern at a time of 30 min using 2 mM ClO_2 and 2 mM MA. We have also investigated the effect of the PVA concentration. A concentration of PVA below 2% does not generate a color change robust enough to visualize the patterns. On the other hand, PVA concentrations above 2% resulted in poor contrast, rendering the gel too dark to effectively visualize the patterns. The recommended I_2 concentration is 3.2 mM, although minor deviations from this concentration do not significantly affect the results.

To further investigate the development of transient Turing patterns in our system, images were captured at regular intervals, displaying the transformation from stripes to spots and the length of time the patterns remained stable (Figure 2). The clock was started upon introduction of the gel into the iodine solution. In the system with 2 mM MA and 2 mM ClO_2 , we found that a distinct and observable transient Turing pattern emerged after roughly 8 min following the start of the experiment and a clear pattern at around 15 min. The pattern fully developed around the 20 min mark; however, as time progressed, the pattern transitioned from stripes to spots. After approximately 50 min, the pattern had a significantly smaller proportion of stripes, and continued to transition to only spots.

In this batch system, the concentrations of reactants change continuously, not only as a result of chemical reactions but also due to their dissipation into the surrounding environment. ClO_2 , MA, and PVA diffuse from the gel into the surrounding iodine solution and iodine diffuses from the solution into the gel. Between $t = 20$ and 50 min and between $t = 60$ and 80 min, there is little change in the pattern structure; however, there is a notable change in the color and saturation of the pattern. Eventually, the reagent concentrations fall outside the optimal ranges required for Turing pattern formation. Consequently, the pattern begins to fade noticeably by approximately $t = 80$ min. The diminishing reagent concentrations disrupt the delicate balance necessary for sustaining the pattern, ultimately resulting in its disappearance.

As seen in Figure 3, experiments conducted with 1 mM MA and 1 mM ClO_2 exhibited a slower onset of the pattern, with a discernible formation beginning after roughly 15 min, almost twice as long as at the higher concentrations. The fully formed pattern was evident after roughly 30 min, and the ratio of stripes to spots was considerably lower in this instance due to the diminished concentration of reagents utilized; any stripes that did form vanished quickly. By altering the concentrations of reagents in the CDIMA system, it is possible to produce

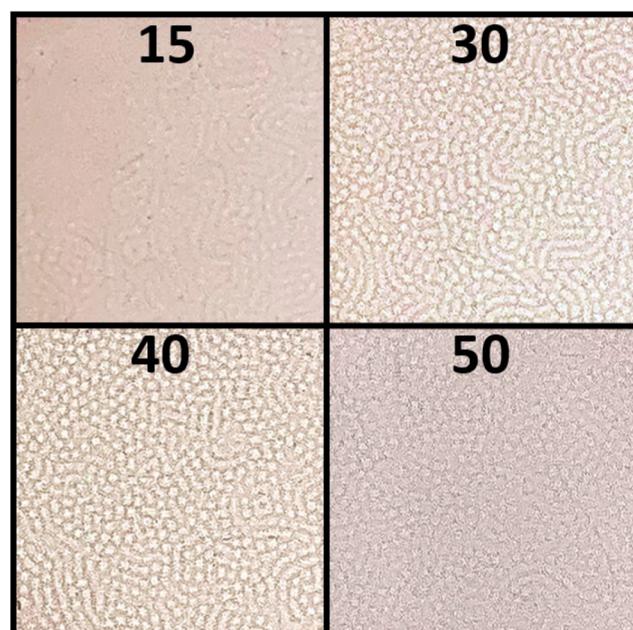


Figure 3. Stability of Turing patterns at later times in a batch system. $[\text{MA}] = 1 \text{ mM}$, $[\text{ClO}_2] = 1 \text{ mM}$. Times (in minutes) are displayed in snapshots, the system size shown is $10 \times 10 \text{ mm}$.

diverse forms of Turing patterns, including spots and stripes, in different ratios. To achieve the most robust pattern formation and clear definition of spots and stripes, it is recommended to utilize 2 mM concentrations of the reagents in demonstrations. However, it is worth noting that pattern formation can still occur at lower concentrations of MA and ClO_2 . While the pattern may be less pronounced or exhibit reduced definition, there is some margin of error in terms of reagent concentrations, providing flexibility and allowing for slight deviations in the concentrations without completely hindering pattern formation. Additionally, the variation in the proportion of spots and stripes can be effectively utilized as an instructional tool for elucidating how alterations in the reaction parameters influence the pattern morphology, providing valuable insights to students.

We also developed another method and setup for generating Turing patterns in the CDIMA reaction in a batch system, which obviates the need for an agarose gel. Our methodology is described in the [Supporting Information](#). While this method of pattern generation may offer a faster reaction time, we conclude that the system with the agarose gel remains superior for demonstrations, as it enables the production of both spots and stripes with more regular spatial periodicity and patterns that are visible for a longer period of time.

Reaction–Diffusion Mechanism and Turing Patterns

Reaction–diffusion (RD) mechanisms constitute an intricate paradigm within the domain of physical chemistry, which is typically expounded upon in advanced courses pertaining to thermodynamics and kinetics. Turing patterns formation in a RD system involves the interplay of an activator and an inhibitor, as demonstrated by Figure 4.^{2–6} This process operates in opposition to the anticipated results based only on simple diffusion. In this scenario, the differential diffusion dynamics of the activator and the inhibitor yield distinct concentration gradients.⁵⁶ Specifically, the activator exhibits a relatively slower diffusion rate toward areas of low activator

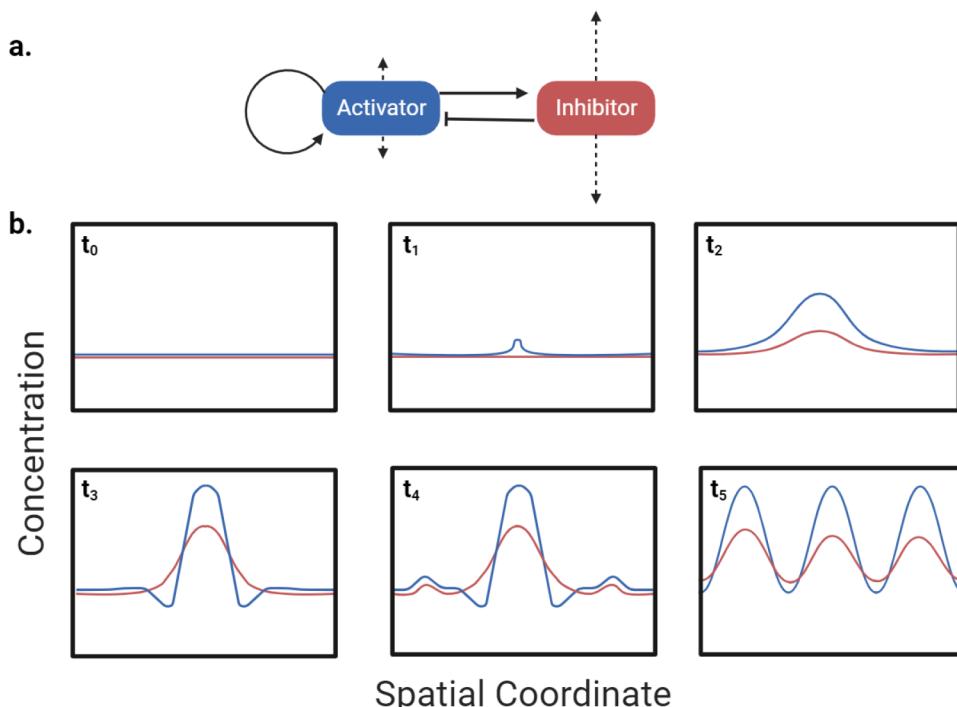


Figure 4. An illustrative elucidation of Turing pattern emergence within a reaction–diffusion system with fast diffusing inhibitor and slow diffusing activator. a) The activator undergoes a self-production mechanism, thereby generating itself through autocatalysis while initiating the production of an inhibitor. This inhibitor subsequently exerts a constraining influence on activator production. The dissimilarity in diffusion rates is depicted through varying lengths of the diffusion arrows (represented by dashed lines). b) A graphical representation of pattern formation within the activator-inhibitor model. At t_0 , the system commences in a state of uniform concentrations of the chemical species across the entire system. At t_1 , after a small perturbation in the activator concentration in the center, an activator production augmentation is initiated in this location. At t_2 , the activator autocatalytic self-production increases its concentration. The heightened activator production simultaneously triggers an increase in inhibitor production in the same location, giving rise to the emergence of a minor peak in inhibitor concentration. At t_3 , the inhibitor exerts a suppressive influence on the activator concentration in the vicinity of the highly concentrated activator, consequently restricting the activator self-production. This sequence of events results in the formation of a prominent peak in activator concentration, encircled by regions characterized by notably lower activator concentration. At t_4 , the activator and inhibitor continue to diffuse across the system with different speeds and activator and inhibitor concentrations form new peaks. At t_5 , a stable state is reached and the concentrations of both the activator and inhibitor adopt a wavelike pattern.

concentration, whereas the inhibitor demonstrates a swifter diffusion toward the same regions, resulting in a discrepancy between their respective spread rates. Consequently, the inhibitor's rapid migration toward the low activator concentration zone leads to a more rapid depletion of the activator concentration through a reaction mechanism, surpassing the rate at which the activator concentration replenishes via diffusion.⁵⁷ This imbalance results in the formation of localized regions characterized by a wavelike pattern of elevated activator concentration juxtaposed against regions marked by diminished activator concentration. The uniform equilibrium state is intrinsically unstable, while the patterned or ordered state attains stability, giving rise to the characteristic Turing patterns.⁶ The Turing pattern demonstration, when employed as an instructional tool in a lecture focused on RD phenomena, affords students a tangible opportunity to directly observe the operational principles underlying such systems and to observe how the CDIMA reaction is a quintessential example of a RD system that embodies the core principles necessary for pattern formation. This demonstration showcases the interplay of autocatalysis, inhibition, diffusion, nonlinearity, and instability, resulting in the spontaneous emergence of spatial patterns.

Classroom Settings

Using this gel-based method for classroom demonstrations is especially advantageous as it provides a straightforward and

convenient means to exhibit the captivating occurrence of pattern formation in chemical systems. The concept of self-organization in chemical systems could be enhanced by this visual experience. We propose three possible classroom settings in which this experiment could be developed:

1. Demonstration for a larger group of students

In a large lecture hall, this demonstration can be conveniently conducted by positioning a document camera above the Petri dish or using a camera with a closeup view and displaying the pattern in real time on a screen. In this setting, the lecturer might make use of other pedagogical tools to provide explanations about the experiment, as the students follow the formation of the patterns.

2. Demonstration for smaller groups

In a small group setting, this demonstration can be conveniently conducted on a light table. Students will have the opportunity to observe pattern formation by the naked eye by looking at the gel inside the Petri dish. The illumination from the light table will enhance the visibility of the pattern and facilitate a clear view for each participant. In this case, students may have the opportunity to manipulate some important variables, e.g., temperature, and extend discussions about the pattern formation process.

3. Experimental activity in physical chemistry laboratory

Considering the variety of chemical and physical concepts that this experiment encompasses, it may also be included as part of the curriculum of a physical chemistry laboratory course. In this case, the students will be responsible for the preparation of the solutions and apparatus and the execution of the experiment. Additional topics and concepts could be explored in this format. For example, pattern formation can be studied for several sets of reagent concentrations. An experiment can be designed to study the transition from Turing patterns to the emergence of chemical waves when increasing the temperature of the ice–water bath surrounding the Petri dish. Other studies may include evaluation of the patterns’ stability when the experiment is carried out in gel and in liquid-based matrices, thereby providing students with a deeper insight into the intricate kinetics governing these reaction systems.

During a lecture on the kinetics of reaction–diffusion systems, this illustrative demonstration was presented to an audience comprising 30 undergraduate and graduate chemistry students. The feedback from these students proved to be overwhelmingly affirmative. The attendees characterized the demonstration as engaging and informative, noting its substantial contribution to their comprehension of how the reaction–diffusion mechanism underlies the formation of patterns. The unanimous sentiment among the students suggested that prior to this lecture, they possessed minimal to no understanding of reaction–diffusion systems. They collectively stressed the indispensability of the visual aid in enabling them to grasp the intricacies of the subject matter. Furthermore, the feedback included a strong recommendation for the incorporation of this demonstration into advanced undergraduate and graduate-level courses in physical chemistry and, more specifically, the field of kinetics.

CONCLUSION

An innovative pattern formation demonstration, which has the potential to cultivate the interests of students in chemistry as well as to enhance their comprehension of nonlinear dynamics and pattern formation in chemical systems, has been introduced. Notably, this demonstration employs a batch system while utilizing relatively inexpensive equipment, rendering it appropriate for classroom presentations. The current study explores the formation of spots and stripes in pattern formation while investigating the impact of time on the patterns.

Researchers have demonstrated that Turing’s model has the capacity to elucidate shapes and patterns found throughout nature. Patterns demonstrated in the CDIMA batch reaction–diffusion system mimic some of the patterns seen in the natural world, drawing attention to connections between chemical dynamics and various other disciplines of science, including ecology, botany, biology, geomorphology, and physiology. Symmetry breaking pattern formation such as RD pattern formation has the potential to explain macroscopic patterns, thereby holding considerable importance in the formation of complex systems and the fields of social science and economics. This underscores the significance of such demonstrations for students and the importance of nonlinear chemical dynamics.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available at <https://pubs.acs.org/doi/10.1021/acs.jchemed.3c01208>.

Procedure to prepare chlorine dioxide stock solution, recommendations for cleanup post reaction, and Turing patterns in a nongel based batch system ([PDF](#))

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Notes

The authors declare no competing financial interest.

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