Turing patterns visualized by index of refraction variations

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Recently Castets et al.\(^1\) discovered stationary chemical patterns of the type that Turing had predicted could form in a reaction–diffusion system. Then Ouyang and Swinney\(^2\) obtained extended two-dimensional Turing patterns and showed that these arose through a bifurcation from a uniform state. Both of these experiments used a chlorite–iodide–malonic acid reaction with starch as an indicator. However, Ouyang\(^3\) and Lengyel et al.\(^4\) have found that starch plays a more complex role than that of a simple indicator in the oscillatory chlorite–iodide–malonic acid reaction. The present experiments demonstrate that Turing patterns can be observed without the use of starch: These patterns are quite similar to those found by Castets et al. Our patterns are rendered visible by variations in the refractive index due to nonuniform swelling of the gel. In contrast, the patterns obtained using starch are rendered visible primarily by spatial variations in the concentration of the I\(^-\)–starch complex.

We first describe the experiment and then present the results. Figure 1 is a diagram of the gel reactor, which is similar in design to one developed by Noszticzius et al.\(^5\) and identical in cross section (1 mm x 3 mm) to the one used by Castets et al.\(^1\) An important difference is that our gel recipe (given in the caption for Fig. 1) contains an eight times greater concentration of cross linking agent\(^6\) than that used by Castets et al.\(^1\)

In a preliminary experiment we used a gel containing starch (2% Thiodene from Prolabo) and reproduced the patterns previously obtained by Castets et al. Then we made a gel that contained no starch and obtained the pattern shown in Fig. 2(a), which closely resembles the patterns obtained with starch. Figure 2(a) was observed in transmitted light of wavelength 352 nm, where I\(^-\) strongly absorbs. When the same pattern was examined at 572 nm, where the I\(^-\) absorption is weak, Fig. 2(b) was obtained: the dark left-hand side of Fig. 2(a) had become light, indicating that this dark region arises from I\(^-\) absorption. Surprisingly, however, the dot pattern remains quite visible, which indicates that the dot pattern does not arise from I\(^-\) absorption.

As a step towards understanding the origin of the patterns that form without starch in the gel, we washed the gel by changing the reactor feeds to pure water. The pattern of white dots faded and disappeared, as expected. However, unexpectedly, the pattern slowly reappeared as black dots on a white background, as shown in Fig. 2(c). This "fossil" of the original chemical pattern indicates that the reaction caused a change in the gel matrix. We then focused our attention on the fossil pattern, which is an equilibrium structure that is easier to study than nonequilibrium reaction–diffusion patterns, which require a continuous feed of chemicals to be sustained.

The fossil patterns correspond to a spatial variation in the refractive index. The dots are regions in the gel with higher or lower index of refraction playing the role of small lenses. This became clear when the camera was moved toward the gel—the black dots in Fig. 2(c) faded and white dots emerged. Similarly the white dots in the original Turing pattern, Fig. 2(a), also arise from a variation in refractive index—the pattern slowly changed to one with black dots as the camera was moved toward the gel; see Fig. 2(d). In contrast, for gels containing starch, a translation of the camera in this way resulted in essentially no change in the observed dot pattern.

The visibility of dot patterns obtained without starch in the gel was found to decrease rapidly with decreasing concentration of the gel cross linking agent, bis-acrylamide. The gel patterns became invisible for 0.1% bis-acrylamide in the gel, which is a concentration one-eighth of that used in obtaining the patterns in Fig. 2. With starch in

![FIG. 1. A schematic diagram of the gel reactor. Sides A and B (each 1 mm x 100 mm) were fed from stirred reservoirs. Reservoir A contained 24 mM NaClO\(_2\) and Reservoir B contained 10 mM malonic acid and 10 mM H\(_2\)SO\(_4\); both reservoirs also contained 3 mM NaI, 3 mM Na\(_2\)SO\(_4\), and 3 mM NaOH. The reservoir residence times were 2 min. The reactor temperature was 2.5 °C. A mixed monomer solution (5 mL of 16% acrylamide and 0.8% bis-acrylamide), together with the initiator (80 μL of 30% triethanolamine and 80 μL of 20% ammonium persulfate), was poured into the cavity between the quartz plates, where the polymerization occurred.](https://ojps.aip.org/jcp/jcpcr.jsp)
FIG. 2. Computer-enhanced video images of sustained stationary patterns in a gel containing no starch, observed in a 0.80 mm × 0.75 mm region in the central region of the reactor. (a) Turing pattern observed in absorption at 352 nm. The distance between the dots is 0.18 mm. (b) Turing pattern observed at 572 nm. (c) Pattern in a gel that has been washed in distilled water, observed at 352 nm. (d) Same as (a), except that the camera was moved closer to the gel.

A gel made with the high bis-acrylamide concentration (0.8%), the observed patterns were found to arise from both absorption and index of refraction variations. This was demonstrated using an optical filter made of a solution of the I−−starch complex: The originally dark region of the reactor (where the I−−starch complex concentration was high) became transparent, but the dot pattern remained visible and its location was unchanged.

The fossil patterns disappeared when the gel was washed in a dilute electrolyte (0.01 to 0.03 M HCl, NaCl, or NaOH). HCl was the most effective in changing the pattern, followed by NaCl and NaOH. These changes were reversible: The original fossil pattern was always recovered when the gel was washed again in pure water. These observations suggest that fixed polar groups in the gel network, most probably acrylic acid groups, play an essential role in the fossil patterns. The uneven distribution of these fixed groups is probably due to an uneven etching of the gel in the presence of the original Turing pattern. Thus the Turing pattern is fixed in the gel, and this fossil pattern arising from osmotic pressure differences appears after the chemicals are removed by washing.

We conclude that, for gels with a high concentration of cross linking agent, there is a nonuniform swelling of the gel when a Turing pattern forms, and the resultant refractive index pattern reappears after the chemicals are removed by washing. The Turing pattern appears to have the same geometry and wavelength for gels with low and high concentrations of cross linking agent and with and without starch. No significant change was observed when the patterns were prepared with high purity monomers (electrophoretic grade acrylamide and bis-acrylamide) or with a different cross linking agent (piperazine diacrylamide). Thus in the absence of starch in a highly cross-linked gel, variations in the refractive index indicate the Turing structure just as variations in absorption indicate the patterns in gels with starch. We cannot rule out the possibility, however, that the swelling of the gel also plays some active role in the formation of the observed patterns.

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6 The swelling of our gel was negligible due to the high concentration of cross linker (0.8% bis-acrylamide) and to the confinement of the gel. Castets et al. (Ref. 1) reported a swelling by a factor of 3 in volume.