Electronic supplementary information

Spatiotemporal generation of alternating disparate pH domains via audible sound controlled opposing enzymatic reactions

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General information

All the reagents and solvents employed were commercially available and used as supplied without further purification. Deionized water with a resistivity of 18.2 MΩ cm^{-1} was used to prepare aqueous solutions. A function generator (AFG-2005, GW Instek) and a speaker (PC83-8, Dayton Audio) were used to generate and control vertical vibrations. Vibrational acceleration was measured by a vibration meter (ST-140, Tenmars). Photos of the experiments were taken by a smartphone.

General procedure for sound induced colored pattern generation experiments

A circular glass Petri dish was mounted on top of a loudspeaker with a flat acrylic tray and the loudspeaker was connected to a function generator to generate vertical sinusoidal vibration. The typical experimental set up is shown in Fig. S1. The ranges of frequency and amplitude of vibration were controlled by a function generator and the amplitude of vibration was measured with a vibration meter. For the audible sound induced transient domains and color pattern generation experiments, 40 Hz with an amplitude of the vibration in the range of 0.20−0.25 g were found to be suitable. A 56 mm-sized (inner diameter) circular glass Petri dish was used for pattern generation experiments. Each pattern generation experiment was repeated more than 10 times to confirm the reproducibility of the pattern formation process.

Spatiotemporal generation of acidic pH domains with glucose oxidation

A 5.0 mL of solution containing glucose (90 mM), bromocresol purple (0.40 mM), and NaCl (0.15 M) in deionized water was purged with argon gas for 0.5 h and mixed with GOx (50 U/mL) and catalase (4.5 kU/mL). The solution pH was then adjusted to neutral with 0.010 M NaOH, and the violet solution was gently poured into a Petri dish placed on a tray. After waiting for several seconds for stabilization, the pattern generation in the dish was recorded with a smartphone.

Generation of basic pH domains with urea hydrolysis

A 5.0 mL of solution containing urea (50 mM), URE (0.50 U/mL), cresol red (0.10 mM), and NaCl (0.15 M) in deionized water was prepared, and the pH of the solution was adjusted to neutral with 0.010 M HCl. The yellow solution was gently poured into a Petri dish placed on a tray, and then the Petri dish was gently shaken to ensure that the solution was homogenous. After waiting for several seconds for stabilization, the pattern generation in the dish was recorded with a smartphone.

Spatiotemporal generation of alternating disparate pH domains with glucose oxidation and urea hydrolysis

A 5.0 mL of solution containing glucose (140 mM), urea (50 mM), bromothymol blue (0.40 mM), and NaCl (0.15 M) in deionized water was purged with argon gas for 0.5 h and mixed with GOx (40 U/mL), URE (0.25 U/mL), and catalase (3.6 kU/mL). The solution pH was then adjusted to neutral with 0.010 M HCl. The green solution was gently poured into a Petri dish placed on a tray and then the Petri dish was gently shaken to ensure that the solution was homogenous. After waiting for several seconds for further stabilization, the pattern generation

in the dish was recorded with a smartphone.

Colored pattern analyses

The images obtained from the bromothymol blue pattern experiments were analyzed using ImageJ software. As the background color is green, using red instead of yellow allows for more accurate analysis. For each image at a particular time, 10 data points were collected from the region of interest. The same work was carried out in a series of ten experiments and similar results were obtained. The percentage change in red was calculated by subtracting the initial red value. The error bars in the line profile figures represent the standard deviation of the pattern image intensity.

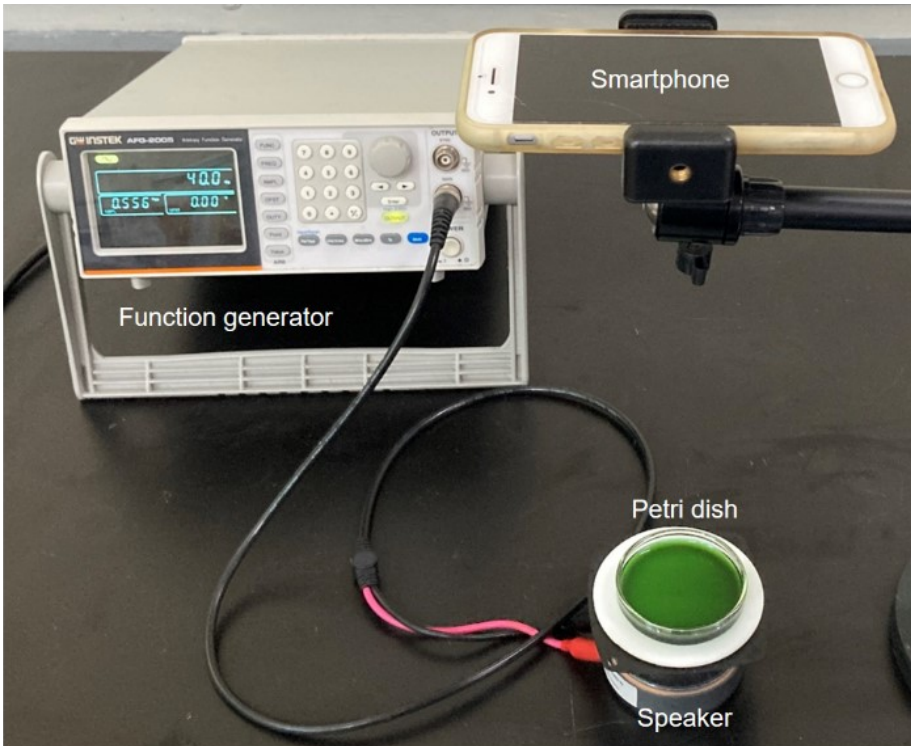

Figure S1. A photograph of the experimental setup.

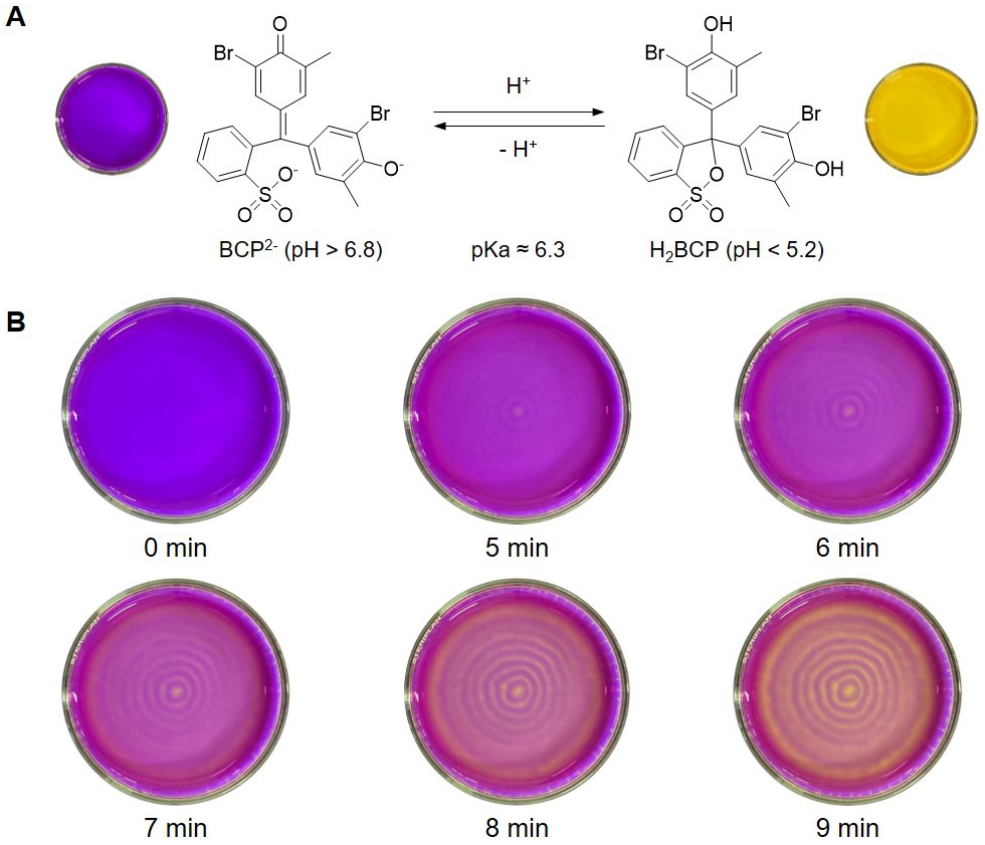

Figure S2. Spatiotemporal generation of alternating acidic pH domains generation with glucose oxidation. (A) Changes in structure and color according to pH change of bromocresol purple (BCP). (B) Time-dependent changes in pattern with 40 Hz sound.

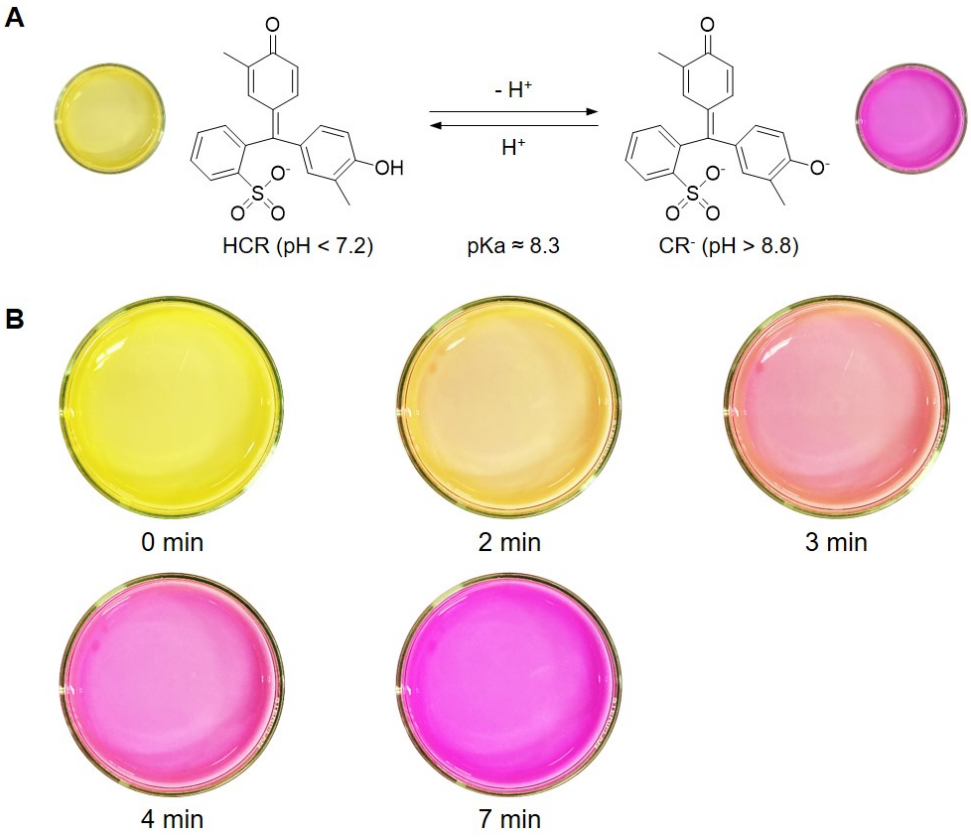

Figure S3. Generation of basic pH domains with urease hydrolysis. (A) Changes in structure and color according to pH change of cresol red (CR). (B) Time-dependent changes in pattern with 40 Hz sound.

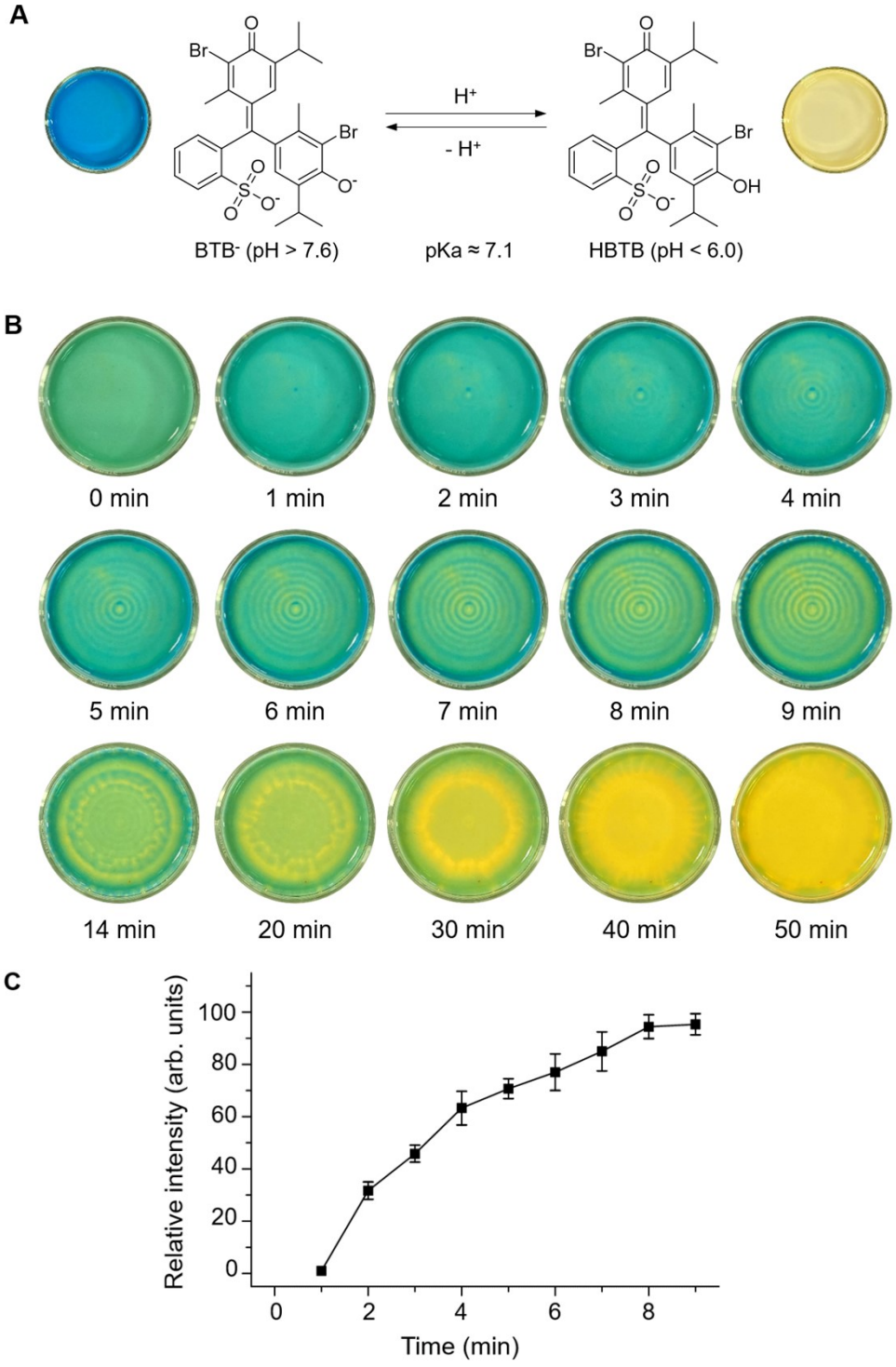

Figure S4. Spatiotemporal generation of alternating disparate pH domains with glucose oxidation and urease hydrolysis. (A) Changes in structure and color according to pH change of bromothymol blue (BTB). (B) Time-dependent changes in pattern with 40 Hz sound. (C) Time-dependent changes in the image color intensity with standard deviation (N = 10) at the central region of the pattern.

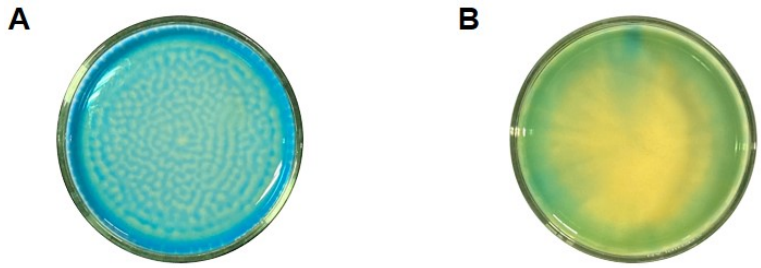

Figure S5. Effect of the sound-induced vibration amplitude on pattern generation with glucose oxidation and urea hydrolysis. (A) Weak vibration (amplitude = 0.1 g). (B) Strong vibration (amplitude = 0.3 g). Each image was taken after 9 minutes of the reaction.

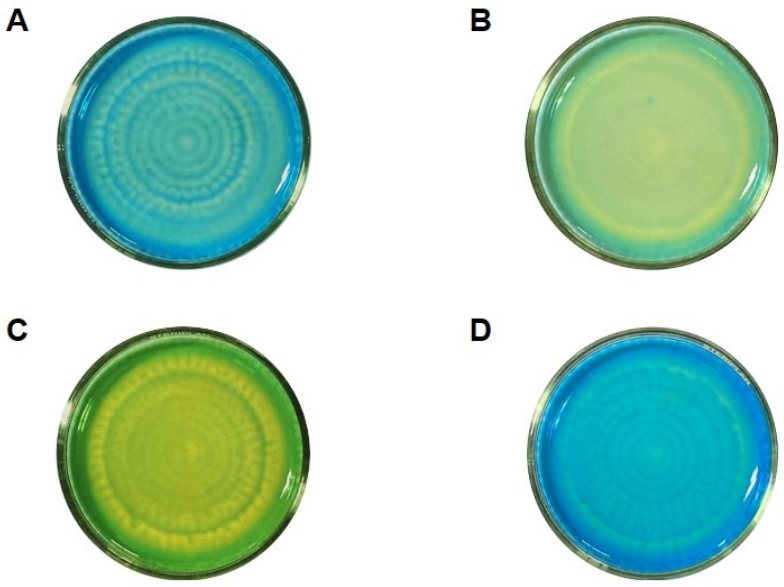

Figure S6. Effect of glucose and urea concentrations on pattern generation with glucose oxidation and urea hydrolysis. (A) Glucose 100 mM, urea 50 mM. (B) Glucose 200 mM, urea 50 mM. (C) Glucose 140 mM, urea 25 mM. (D) Glucose 140 mM, urea 75 mM. With the exception of the glucose and urea concentrations, the other conditions are the same throughout the experiment. Each image was taken after 9 minutes of the reaction.