Dynamics of spiral waves and bubble formation in a closed chemical system of thin photosensitive excitable media

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Abstract. Spiral waves have been observed in a thin layer of excitable media. Especially, electrical spiral waves in cardiac tissues connect to cardiac tachycardia and life-threatening fibrillations. The Belousov-Zhabotinsky (BZ) reaction is the most widely used system to study the dynamics of spiral waves in experiments. When the light sensitive $Ru(bpy)_3^{2+}$ is used as the catalyst, the BZ reaction becomes photosensitive and the excitability of the reaction can be controlled by varying the illumination intensity. However, the typical photosensitive BZ reaction produces many CO_2 bubbles so the spiral waves are always studied in thin layer media with opened top surfaces to release the bubbles. In this work, we develop new chemical recipes of the photosensitive BZ reaction which produces less bubbles. To observe the production of bubbles, we investigate the dynamics of spiral waves in a closed thin layer system. The results show that both the speed of spiral waves and the number of bubbles increase with the concentration of sulfuric acid (H_2SO_4) and sodium bromate ($NaBrO_3$). For high initial concentrations of both reactants, the size of bubbles increases with time until the wave structures are destroyed. We expect that the chemical recipes reported here can be used to study complicated dynamics of three-dimensional spiral waves in thick BZ media where the bubbles cannot escape.

1. Introduction

Spiral waves have been found in a thin layer of different excitable media, including CO oxidation on a platinum surface [1], the slime mold Dictyostelium discoideum [2], cardiac tissue [3], and the Belousov-Zhabotinsky (BZ) reaction [4]. Such spiral patterns of an electrical signal (action potential) in heart tissues can occur that also have an important effect on human health. It is found some cardiac arrhythmia (e.g., atrial and ventricular tachycardia [5]). Questionability of the spiral waves pattern expanded into waves turbulence [6] which corresponded to ventricular fibrillation [5] and can lead to a sudden cardiac death. The waves turbulence can be investigated only in a three-dimensional (3D) system. It has observed in anisotropic myocardium tissues with thickness of 3-10 mm [6].

In very different systems, spiral waves are governed by a reaction-diffusion mechanism [7]. The BZ reaction is the most popular reaction to study the dynamics of spiral waves because it is easy to prepare and convenient to detect the wave structures. Moreover, the system can be easily controlled by the light intensity when the light sensitive catalyst ($Ru(bpy)_3^{2+}$) is filled in the BZ reaction. The control of spiral waves by light are reviewed in [8].

The mechanism of the BZ reaction is very complex and involve many different steps. The overall reaction: $3\ CH_2(COOH)_2 + 4\ BrO_3^- \rightarrow 4\ Br^- + 9\ CO_2 + 6\ H_2O$ shows CO_2 as one of the end products [9].

In the course of time, CO_2 increases and appear as bubbles in close unstirred BZ systems. The dynamics of spiral waves is disturbed when the bubbles are large and act as unexcitable defects [10]. The occurrence of CO_2 bubbles is an important problem in the BZ reaction, especially in a thick media. The effect of CO_2 bubbles not only perturbs the structure of spiral waves but also obstruct observations of 3D structures [11]. The typical photosensitive BZ reaction usually [12-15] use a high initial concentration which produce plenty bubbles. Therefore, photosensitive spiral waves are studied in thin layer media with opened top surfaces to release the bubbles.

In this research, we modify the chemical recipes of photosensitive BZ recipes to reduce the bubble production. We use agarose to reduce the hydrodynamic effect and observe them in a closed thin layer system to study the dynamics of spiral waves.

2. Methods

In all experiments, 10 ml of the photosensitive BZ solutions are prepared from $NaBrO_3$, H_2SO_4 and Malonic acid (MA) all purchased from Merck. Moreover, we use Tris(bipyridine)ruthenium (II) chloride ($Ru(bpy)_3Cl_2$) as a catalyst (Sigma). Stock solutions of $NaBrO_3$ (1M), $Ru(bpy)_3Cl_2$ (25mM) and MA (1M), are prepared by dissolving the chemicals in deionized water (conductivity ~ 0.056 $\mu S\ cm^{-1}$) while stock solution of H_2SO_4 (2.5 M) is purchased. To prevent the hydrodynamic perturbation, the reaction is embedded in a 1.0 % w/w agarose gel (Sigma). The BZ solution is prepared with a heat solution of agarose then the temperature is allowed to reduce until it reaches 36 °C and finally the reactants is filled. These solutions are prepared with different initial reagent concentrations: [$NaBrO_3$] = 0.05-0.20 M, and [H_2SO_4] = 0.4 – 1 M, whereas [MA] = 0.05 M and [$Ru(bpy)_3Cl_2$] = 0.625 mM. In addition, a surfactant, sodium dodecyl sulfate (SDS, Fluka), is added to the solution to decrease CO_2 bubble production [9].

Dynamics of spiral waves in photosensitive BZ solution are investigated in a flat closed reactor constructed from transparent Plexiglas. After the BZ solution is filled into the reactor with 60 x 60 x 0.6 mm^3 of volume, propagating waves occur spontaneously, usually at the boundary of the reactor. We shine a strong spotlight (power =150 watts, diameter = 5 mm) to a segment of wave front so its propagation is entirely suppressed. After that those two free ends of fronts start curling and develop to spiral waves.

The reactor is placed between a projector and a color CCD camera (Super-HAD, Sony) to record the images every second with a resolution of 0.03 mm $pixel^{-1}$. All positions of the reactor are homogeneously illuminated with 350-lux blue light (a blue image with RGB value of [0 0 255]) using a video projector (BenQ XGA). Note that the projector and the light beam produce heat that causes the temperature of the reaction increases in the absence of cooling system. Therefore, we carefully control the temperature of the reactor by using an air conditioner and a fan blowing air direct to the reactor. After a transient interval at the beginning of experiments, the temperature of the reactor is well controlled to 22 ± 1 °C.

3. Results

Figure 1 shows examples of the spiral waves in the photosensitive BZ reaction with [$NaBrO_3$] in the range of 0.05-0.20 M whereas [H_2SO_4] = 0.4 M and the concentrations of the other reagents are described in the methods. Spontaneous waves occur and the spiral wave generation can be started shortly after the solution is poured into the reactor, except for the lowest [$NaBrO_3$] in figure 1(a), they appear about 10 minutes slower than the others.

In cases of low concentrations, [$NaBrO_3$] ≤ 0.1 M, the media look clear without CO_2 bubbles from the beginning [figures 1(a) and 1(b)] until the end of the experiments [figures 1(a′) and 1(b′)]. In contrast, tiny bubbles appear in those with higher concentrations, [$NaBrO_3$] ≥ 0.15 M [figures 1(c) and 1(d)]. In the course of time, the size and amount of such bubbles increases and disrupt the wave structures [figures 1(c′) and 1(d′)]. It is worth noting that we also observe appearances of the bubbles and the disruption of the waves in the other series of experiments with [H_2SO_4] ≥ 0.70 M where [$NaBrO_3$] is kept constant at 0.05 M (images not shown).

For all spiral waves in this study, their shapes are unchanged after they evolve about 10 rotations, so we start to analyse their dynamics at this state. The spiral tips at the center of the wave structures rotate around small circles namely the spiral cores. Far from the spiral cores, the period *T*, wavelength λ, average speed *v* (*v* = λ/T) of the wave fronts are measured.

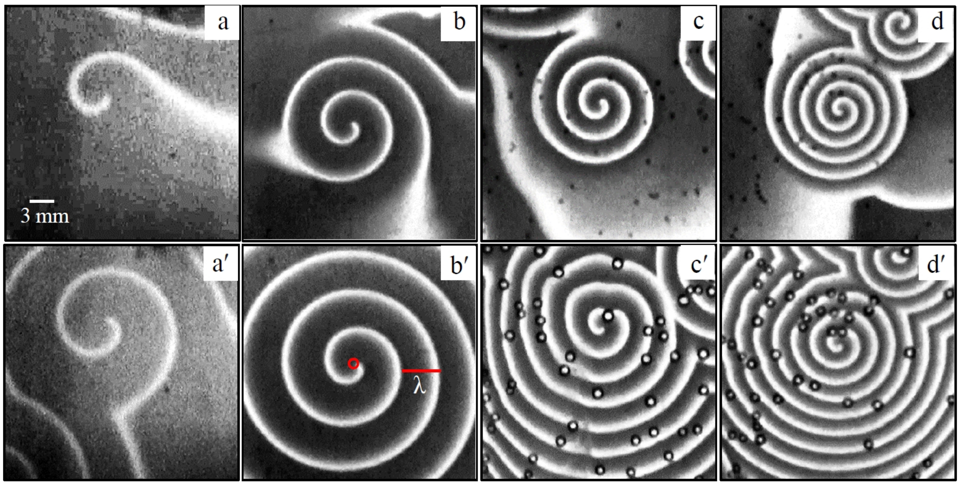

Figure 1. Spiral waves in the photosensitive BZ reaction with different [$NaBrO_3$]: (a, a′) 0.05, (b, b′) 0.1, (c, c′) 0.15, and (d, d′) 0.2 M. Top and bottom rows show the spiral waves after about 10 and 30 rotations, respectively. The other initial concentrations are [H_2SO_4] = 0.4 M, [MA] = 0.05 M and [$Ru(bpy)_3Cl_2$] = 0.625 mM. In (b′), The small circle represents the spiral core and the wavelength λ is depicted.

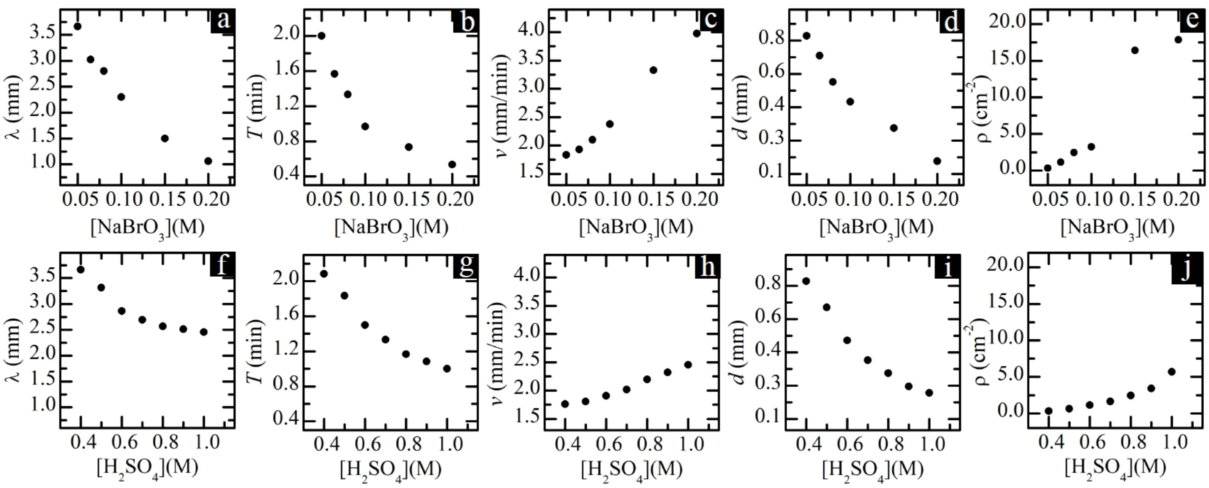

Figure 2. The dynamics of spiral waves in the photosensitive BZ reaction and the density of CO_2 bubbles with a variation of (a-e) [$NaBrO_3$] or (f-j) [H_2SO_4]. The standard concentrations are [$NaBrO_3$] = 0.05 M, [H_2SO_4] =0.4 M, [MA] = 0.05 M and [$Ru(bpy)_3Cl_2$] = 0.625 mM.

The analysis of the dynamics of spiral waves and the density of CO_2 bubbles are summarized in figure 2. They are divided into two series of experiments with a variation of [$NaBrO_3$] or [H_2SO_4]. Figures 2(a) – 2(e) show that when [$NaBrO_3$] is increased, the period *T*, wavelength λ and the spiral core diameter *d* decrease but the average speed *v* increases. As shown in figures 2(f) – 2(i), the effect of [H_2SO_4] on the dynamics of spiral waves is similar to that of [$NaBrO_3$], i.e., when [H_2SO_4] is increased, the period *T*, wavelength λ and the spiral core diameter *d* decrease but the average speed *v* increases.

To compare CO_2 bubbles in different experiments, we measure the density ρ of CO_2 bubbles in the images during 20-30 wave periods where the bubbles have a similar size of about 1 mm (e.g., as in figures c′ and d′). The number n of such bubbles in the observed area A of 6.15 cm^2 is counted and the density ρ is calculated as $\rho = n/A$. Figures 2(e) and 2(j) show that the density ρ of CO_2 bubbles increases with [$NaBrO_3$] and [H_2SO_4], especially it overshoots when [$NaBrO_3$] reaches 0.15 M.

4. Conclusion and discussion

We have presented an investigation of the dynamics of spiral waves in a closed thin layer of the photosensitive BZ reaction with new chemical recipes to reduce the production of CO_2 bubbles. For low concentrations of $NaBrO_3$ and H_2SO_4, the reaction has very few CO_2 bubbles. In contrast, for high concentrations of these reagents, the amount and size of bubbles increase with time. The solutions with chemical recipes in this study support rigidly rotating spiral waves whose tips rotate around circular spiral cores. When the concentration of $NaBrO_3$ or H_2SO_4 is increased, the period, wavelength and spiral core diameter decrease but the wave speed increases.

Traditionally, experiments have been done using the photosensitive BZ reaction with [$NaBrO_3$] = 0.2 - 0.55 M, [H_2SO_4] = 0.3 – 0.77 M, and [MA] = 0.17 – 0.3M [11-14]. The reactions with such high concentrations of reagents produce plenty of CO_2 bubbles so the investigations were limited to perform using thin layer media with opened top surface which can release the bubbles to the atmosphere.

In our study, the experiments are intentionally performed using the photosensitive BZ solutions with closed top surface to observe the bubble production in the course of time. The results suggest that the reaction with low initial concentrations of $NaBrO_3$ and H_2SO_4, produces low bubble density (see figures 2e and 2j) so we expect that some chemical recipes reported here are suitable for a study of 3D spiral waves in thick BZ media, where complicated and exotic wave dynamics can occur. We plan to perform 3D experiments in the future.

Acknowledgment

We thank the Research and Development Institute (KURDI), Kasetsart University, the Thailand Research Fund (Grant No. RSA6280088) and the National Research Council of Thailand (Grant No. NRCT5-RGJ63002-029) for financial support.

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