Chemical Physics Letters 555 (2013) 300–305

Contents lists available at SciVerse ScienceDirect

Chemical Physics Letters

journal homepage: www.elsevier.com/locate/cplett

Synchronization of oscillatory chemiluminescence with pulsed light irradiation

Shunsuke Takayama, Kunihiko Okano, Kouichi Asakura *

Department of Applied Chemistry, Faculty of Science and Technology, Keio University, 3-14-1, Hiyoshi, Kohoku, Yokohama 223-8522, Japan

ARTICLE INFO

Article history:
Received 18 September 2012
In final form 7 November 2012
Available online 16 November 2012

ABSTRACT

A chemical oscillator, the H_2O_2–KSCN–$CuSO_4$–NaOH system, generates an oscillatory chemiluminescence when luminol is added to this system. Attempts were made to synchronize the oscillatory chemiluminescence with pulsed light irradiation. A period of the chemical oscillation became shorter by the irradiation of white and blue color light, while the oscillatory behavior was scarcely influenced by the irradiation of red light. Pulsed red and white or blue lights were irradiated on either the non-luminol or luminol-added H_2O_2–KSCN–$CuSO_4$–NaOH system. Synchronization of the chemical oscillation was achieved for 25–30 min in the luminol-added system.

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1. Introduction

Temporary periodic behaviors, such as circadian rhythm in a cell cycle and cardiac pulsation, are generated in living systems. Besides living systems, artificial chemical systems such as Bray–Liebhafsky (BL) reaction [1,2] and Belousov–Zhabotinsky (BZ) reaction [3] also exhibit temporary periodic patterns called chemical oscillation. In these chemical systems, temporally periodic variations in concentrations of chemical species are observed. Chemical oscillations are the examples of dissipative structures, i.e., self-organization in far-from-equilibrium systems [4–6].

The oscillatory behaviors of the chemical systems are thus influenced by the condition of outer environment; coupling and synchronization of the oscillations are observed when two or more chemical oscillators interact with one another. Two BZ oscillators were found to be coupled by matter exchange through a perforated wall [7] or an aperture of variable sizes [8]. They also exhibit chemical chaos, entrainment, rhythmogenesis, and phase death [9]. In addition, coupling and synchronization was found in electrically connected BZ oscillators [10]. An oscillatory combustion seen in a burning candle is another attractive example of chemical oscillation, and coupling and synchronization were also observed in this system [11].

There is a biological oscillatory system in nature that synchronizes with each other via optical coupling. Fireflies exhibit temporary periodic chemiluminescence, and a fascinating synchronous flashing of fireflies can be observed when they gather in one tree [12]. This phenomenon can be regarded as synchronization of biological oscillations via optical coupling. The present Letter attempts to establish the chemical system that exhibits oscillatory chemiluminescence synchronized with pulsed light irradiation.

Effects of photo-irradiation on temporal, spatial, and spatiotemporal patterns in chemical systems placed far from equilibria have been studied extensively. In a classic BL reaction, oscillation was found to occur more frequently by illumination on reactive solutions with relatively low concentrations, while inhibition of the oscillations was observed in higher concentrations [13]. In a $Ru(bpy)_3^{2+}$ catalyzed BZ reactions, for instance, irradiation of visible light was found to inhibit the chemical oscillation [14]. In this case, $Ru(bpy)_3^{2+}$ changed its reactivity by the illumination to accelerate the production of Br^- known as an inhibitor of oscillation [14–19]. However, no study has yet been conducted on analyzing the influence of photo-irradiation on the H_2O_2–KSCN–$CuSO_4$–NaOH chemical oscillator, hereafter abbreviated as HKCN.

This system is known to show oscillatory behavior in color, redox potential, and dissolved oxygen concentration [20]. Coupling of two subsystems, H_2O_2–$CuSO_4$ and H_2O_2–KSCN, through various intermediates to construct network leads to the oscillatory behavior [21]. The addition of luminol into the HKCN system generates temporally periodic chemiluminescence [22,23]. Since the key intermediate of this system, copper-peroxide complex HO_2–Cu(I), absorbs visible light, the oscillatory behavior is expected to be influenced by the illumination of visible light.

We have here analyzed the influence of photo-irradiation on the behavior of this HKCN system in the absence and presence of luminol. We have also applied pulsed light irradiation to this chemical oscillator to observe its response.

2. Experimental

2.1. Reactions of H_2O_2–KSCN–$CuSO_4$–NaOH

The reactions of HKCN were carried out by mixing aqueous solutions of $CuSO_4$, KSCN, NaOH, and H_2O_2. The concentration of the solution was 0.075, 25, 25, and 250 mM, respectively, and

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* Corresponding author. Fax: +81 45 566 1560.
E-mail address: asakura@applc.keio.ac.jp (K. Asakura).

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http://dx.doi.org/10.1016/j.cplett.2012.11.014

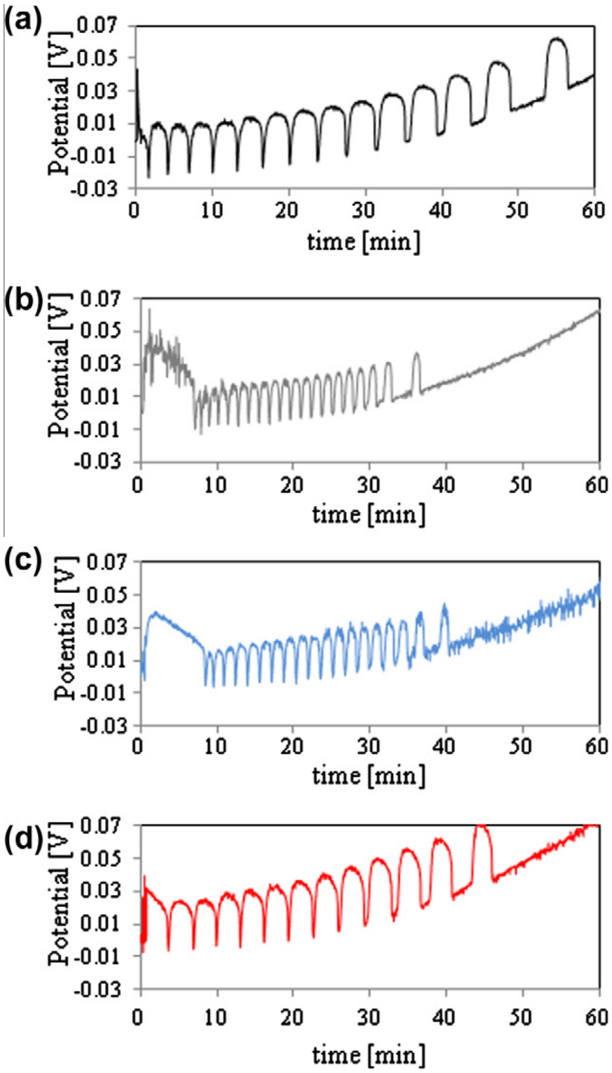

Figure 1. Influence of photo-irradiation on the oscillatory behavior of the HKCN system. Reaction was carried out under the irradiation of (a) no light, (b) white color light, (c) blue color light, and (d) red color light. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

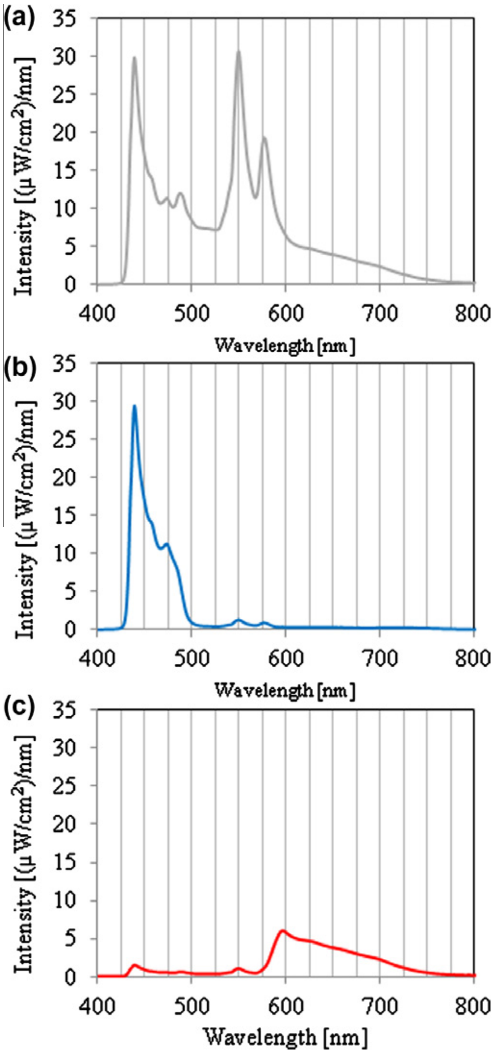

Figure 2. Wavelength distributions of (a) white, (b) blue, and (c) red color light for the photo-irradiation at the reaction flask in the water bath. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

10 mL of each solution was mixed in a 50 mL round-bottom flask at 25 °C with stirring at 300 rpm using a magnetic stir bar (6 mmϕ × 15 mm). The oscillatory behavior was analyzed by a Pt electrode (Hiranuma Sangyo Corporation, PR-701B). In the case of adding luminol into the reaction system, 10 mL of 1.0 mM aqueous solution of luminol containing 25 mM NaOH was mixed with other solutions.

2.2. Photo-irradiation

A projector (Seiko Epson, LCD Projector EMP-1700) was used as a light source for photo-irradiation, and the flask was placed in a water bath to prevent the rise in temperature in the reaction system. The water bath was wrapped with aluminum foil in order to prevent the leakage of light. White, red, and blue color lights controlled by Microsoft Power Point were projected. The radius of the water bath was 9.0 cm, and the distance between the flask and the projector was 15 cm. Wavelength distributions of the lights at the reaction flask in the water bath were determined by a spectroradiometer (Ushio, USR-45). The intensity of the light

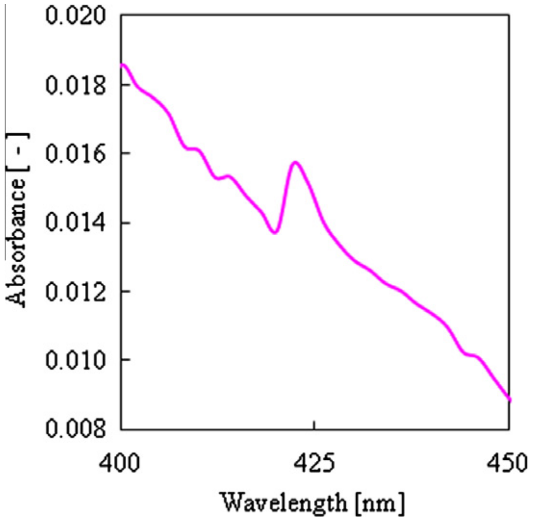

Figure 3. Absorption spectrum of the HKCN reaction solution. The measurement was conducted when the solution was colored during oscillation.

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