# High-resolution and high-speed chemical imaging sensor based on optical fiber array

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# 1. Introduction

The chemical imaging sensor [1, 2] is a semiconductor-based chemical sensor that can visualize the two-dimensional distribution of chemical species. It is based on the principle of the light-addressable potentiometric sensor (LAPS) [3], in which the potential on the sensing surface affects the carrier distribution in semiconductor by field effect. The chemical imaging sensor is expected to be a powerful tool in the fields of electrochemistry, analytical chemistry and biology as well as in the development of microfluidic devices for  $\mu$ TAS.

The conventional systems, however, required a rather long time of measurement to acquire a chemical image, due to the slow mechanical scan of a light beam, which was used to read out the potential distribution on the sensing surface in a pixel-by-pixel manner. In our recent reports [4, 5], we used a two-dimensional array of light source and frequency-division multiplexing for parallel measurement of pixels without the need of slow mechanical scan. We realized spatiotemporal recording of pH images at a high frame rate of 70 fps. In these systems, however, the density of the measurement spots was rather limited due to the dimensions of LEDs. The size and the pitch of LEDs in an array was typically 2.0 mm and 2.5 mm, respectively, which defined the density of measurement spots.

In this study, we propose the use of an optical fiber array as a high-density light source for the chemical imaging sensor system. By using light beams guided by optical fibers, measurement spots on the sensing surface can be defined at much higher densities in spite of the LED size. Combined with frequency-division multiplexing, a high-resolution and high-speed chemical imaging system can be realized. A high-resolution imaging was demonstrated by visualizing the spatiotemporal change of pH distribution during diffusion of enzymatic product.

# 2. Experimental

#### Measurement system

Figure 1(a) shows a schematic view of the high-speed chemical imaging system developed in our previous study. The system consists of a sensor plate, a two-dimensional array of  $8 \times 8$  LEDs as a light source, a measurement PC, and 8-channel oscillator array for frequency-division multiplexing. The sensor plate was made of n-type Si covered with SiO<sub>2</sub> and Si<sub>3</sub>N<sub>4</sub> as a pH-sensitive layer.

For acquisition of a chemical image, 8 LEDs on the same line are turned on at a time, and they are modulated at different frequencies. A light beam from each LED defines a measurement spot and generates a photocurrent signal with amplitude dependent on the surface potential at each position. The externally recorded current signal is a superposition of 8 photocurrent components with respective frequencies, which can be separated by Fourier analysis. In this way, pH values at 8 measurement spots on a line can be determined simultaneously. By repeating the measurement for the 8 lines of the array, a two-dimensional map of pH distribution can be obtained without a mechanical scan.



Fig.1 (a) A high-speed chemical imaging system developed in our previous study. A two-dimensional array of LEDs and frequency-division multiplexing allowed high-speed imaging. However, the density of measurement spots was limited due to the LED size. (b) A high-density light source with optical fiber array proposed in this study, which allows both high-resolution and high-speed imaging. (c) Head part of the optical fiber array.

## *High-density light source*

A two-dimensional LED array was fabricated using  $8 \times 8$  shell-shaped LEDs (Linkman, L3-EKR2530-12500, 17500 mcd). Optical fibers with a diameter of 500 µm (Mitsubishi Rayon, CK-20) were connected to each LED via home-made connecters as shown in Fig. 1(b). Finally, the light-emitting ends of optical fibers were bundled again into an  $8 \times 8$  array with a pitch of 700 µm using a home-made fiber head shown in Fig. 1(c). The optical fiber array used in this study can define measurement spots at a density of about 2 pixels / mm<sup>2</sup>, which is 12 times higher than that defined by the LED array with a pitch of 2.5 mm used in our previous study.

# Observation of enzymatic product

To demonstrate high-resolution imaging, diffusion of enzymatic product was visualized. As shown in Fig.2(a), urease was immobilized at the center of the sensing surface by the following procedure: (1) Mix the albumin solution (Sigma-Aldrich, 10mg/ml, 80µl) and the urease solution (Sigma-Aldrich, 0.1mg/ml, 20µl) in a tube. (2) Add the glutaraldehyde solution (Wako, 25%, 2µl) and mix again quickly. (3) Drop the mixture onto the sensor surface by pipette.



(C)

Fig. 2 (a) Schematic view of the experiment. (b) A series of pH images obtained with a conventional LED array.(c) High-resolution images obtained with an optical fiber array.

The measurement cell was filled with PBS and a urea solution (Sigma-Aldrich, 100mM) was dropped onto urease by a pipette. Urease catalyzed hydrolysis of urea and produced  $NH_4$ . Diffusion of the enzymatic product,  $NH_4$  in this case, was recorded by acquiring pH images repeatedly at a frame rate of 10 fps.

#### 3. Results

Two series of pH images were collected by the chemical imaging system using a conventional LED array and an optical fiber array, respectively. In both cases, increase of local pH indicated production of  $NH_4$ .

Figure 2(b) shows a series of pH images obtained with a conventional LED array. The size of the measured area was  $20.3 \times 20.3 \text{ mm}^2$ , and the area modified with urease was a circle with a diameter of 8mm. In this observation, the whole area modified with urease was monitored with a low density of measurement spots.

Figure 2(c), on the other hand, shows a series of pH images obtained with an optical fiber array. This time, the measured area was  $5.6 \times 5.6 \text{ mm}^2$ , which did not cover the whole area modified with urease. More detailed profile of pH distribution during the diffusion of NH<sub>4</sub> was monitored at a higher resolution.

These results suggest that the use of an optical fiber array as a high-density light source is a promising approach to realize both high-resolution and high-speed chemical imaging sensor system for spatiotemporal recording of chemical dynamics.

### 4. Conclusions

An optical fiber array was employed to define measurement spots of the chemical imaging sensor at a higher density. Combined with frequency-division multiplexing, this approach is expected to realize both high-resolution and high-speed chemical imaging sensor system.

#### References

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