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High-speed Chemical Imaging System Based on Front-side-illuminated LAPS

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Abstract

The chemical imaging sensor is a Si-based chemical sensor that visualizes the spatial distribution of a specific ion in the solution contacted with the sensing surface. It is based on the principle of the light-addressable potentiometric sensor (LAPS), in which a modulated light beam is used to read out the ion concentration in the form of photocurrent. Since the conventional chemical imaging sensor uses a scanning light beam for mapping the distribution of the ion concentration, the long scan time has been a problem for imaging in real time. For high-speed imaging, a plurality of light beams modulated at different frequencies can be employed to measure the ion concentrations simultaneously at different locations. Although a wide frequency bandwidth is required for high-speed imaging, the modulation frequency is limited due to the low-pass characteristics of carrier diffusion in the conventional setup, in which the back-side of the Si substrate is illuminated. In this study, a high-speed chemical imaging system based on front-side-illuminated LAPS was developed, which achieved imaging of pH distribution at 70 frames per second.

Key words: light-addressable potentiometric sensor (LAPS), chemical imaging sensor, EIS, and pH

1. Introduction

The light-addressable potentiometric sensor (LAPS) [1] is a Si-based chemical sensor with an electrolyte-insulator-semiconductor (EIS) structure, in which the Si substrate is illuminated with a modulated light and the induced AC photocurrent is measured to determine the ion concentration on the sensing surface. Here, the measurement area can be defined by illumination. The chemical imaging sensor [2] is based on the addressability of LAPS, in which a scanning light beam is used to obtain a 2-D map of the ion concentration, but it was difficult to observe the dynamics of chemical reaction in real time because of the slow scan rate.

For high-speed imaging, a plurality of light beams modulated at different frequencies can be employed to read out the ion concentrations at different locations in parallel [3, 4] as shown in Fig.1. In this case, the photocurrent signal is a superposition of all frequency components, which can be separated by Fourier analysis. Here, the required bandwidth of the modulation frequency is given by (number of light beams) x (frequency spacing) and the time window of Fourier analysis must be at least as long as 1/ (frequency spacing). This means that high-speed imaging requires a wide bandwidth of the modulation frequency. Due to the low-pass characteristics of carrier diffusion across the sensor plate, however, the modulation frequency is limited in the conventional setup, where the back-side of the Si substrate is illuminated. For front-side illumination, on the other hand, photocarriers are generated and separated directly inside the depletion layer and higher frequencies can be used [5]. In this study, a high-speed chemical imaging system based on front-side-illuminated LAPS was developed using a 2-D array of LEDs.

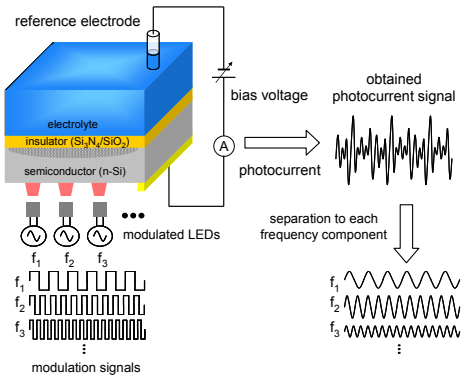

Fig.1 Diagram of simultaneous measurement.

2. Front-side vs. Back-side Illumination

Two different geometries are possible in the measurement of LAPS. In the front-side and back-side illumination, the front-side or the back-side of the Si substrate is illuminated by the modulated light, respectively [5].

As shown in Fig.2 (a), the photocurrent generated by back-side illumination is smaller than that of front-side illumination, due to recombination of photocarriers in the course of diffusion across the sensor plate.

In addition, the decrease of photocurrent with the modulation frequency in back-side illumination is steeper than that of front-side illumination as shown in Fig.2 (b).

This result suggests that front-side illumination is more advantageous for high-speed chemical imaging, for which higher modulation frequencies are required.

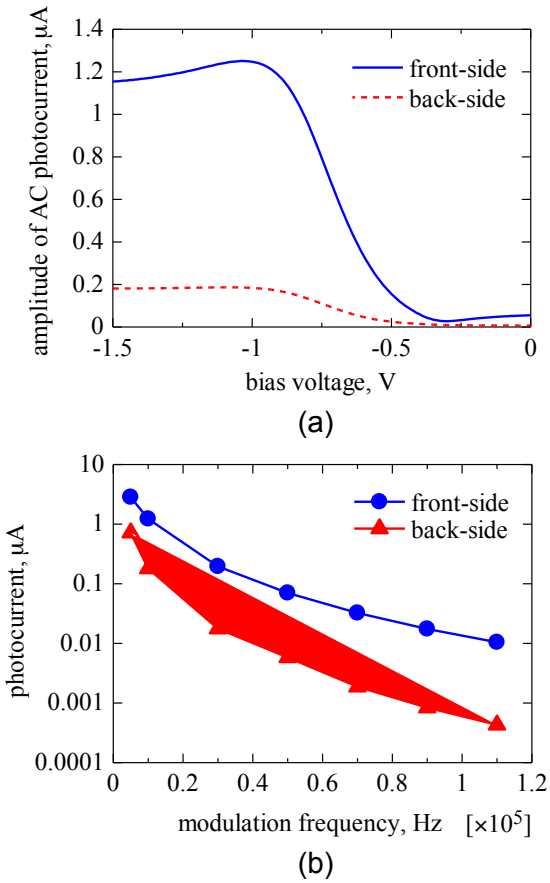

Fig.2 (a) I-V curves for front-side and back-side illumination (LED light modulated at 10 kHz), and (b) magnitude of photocurrent as a function of modulation frequency for front-side and back-side illumination.

3. High-speed Chemical Imaging System

Fig.3 (a) shows a schematic view of the high-speed chemical imaging system based on front-side-illuminated LAPS. The system consists of a sensor plate, an oscillator array to synthesize a series of modulation frequencies, a 2-D array of LEDs (LED matrix) and a control PC.

Sensor plate: The sensor plate is an n-type Si wafer. The top surface of the sensor plate was covered with Si_3N_4 as a pH-sensitive layer on the insulating layer of SiO_2.

Oscillator array: Five modulation frequencies (6, 7, 8, 9 and 10 kHz) are generated by 5-channel phase-locked loop (PLL) synthesizer.

LED matrix: Fig.3 (b) shows a top view of the LED matrix. The matrix has 35 (5×7) LEDs with a separation of 2.5 mm. Five LEDs on one line are modulated at different frequencies, and five points on the sensor surface are measured simultaneously as shown in Fig.4.

PC and Software: The program controls the bias voltage, selects a line of the LED matrix in sequence, measures the photocurrent signal, calculates the amplitude of the signal at each frequency and generates a chemical image.

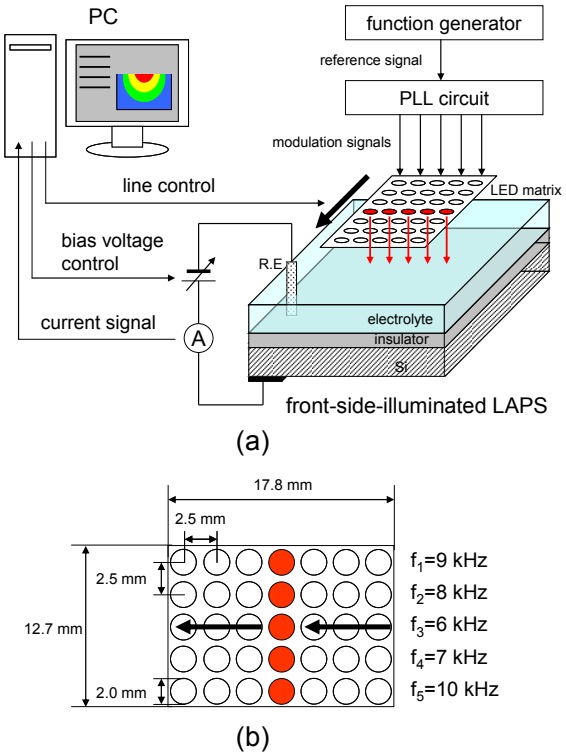

Fig.3 (a) Schematic view of the high-speed chemical imaging system based on front-side-illuminated LAPS, and (b) top view of the LED matrix.

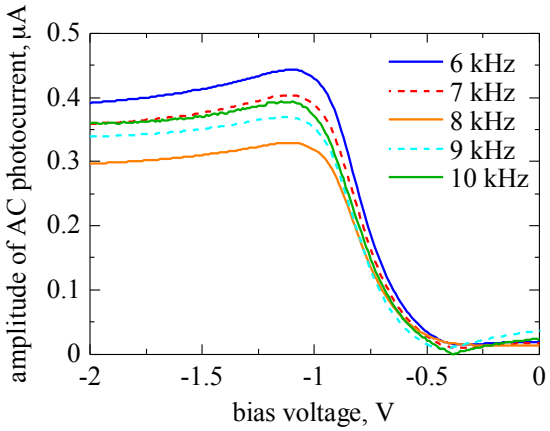

Fig.4 I-V curves for simultaneous measurement on one line of the LED matrix.

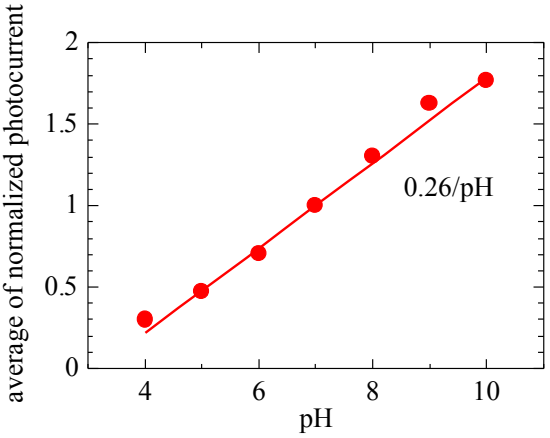

Fig.5 Average of normalized photocurrent as a function of pH value.

4. Experimental Results of Chemical Imaging

Using the developed chemical imaging system based on front-side-illuminated LAPS, the experiments of chemical imaging were performed to observe the temporal change of pH distribution caused by injection of a solution.

In each measurement, the photocurrent values at each measurement position are normalized by the values in the initial state to compensate for the frequency-dependence of the photocurrent and the non-uniformity of the sensor plate.

4.1. Imaging of pH Buffer Solution

Chemical imaging of buffer solutions of pH4, 5, 6, 7, 8, 9, and 10 was performed. The photocurrent values at each measurement position were normalized by the values of pH7 buffer solution. Fig.5 shows an average of normalized photocurrent at each measurement position as a function of pH value. It proves that the normalized photocurrent varies almost linearly with pH value. Fig.6 shows the experimental result of chemical imaging. As shown in Fig.6, the chemical images of each pH buffer solution are almost uniform in color display corresponding to the uniform pH distribution.

4.2. High-speed Imaging at 14 fps

Fig.7 shows the experimental result of high-speed chemical imaging at 14 fps when 0.2 ml of 0.1M HCl solution was injected into 2.0 ml of 0.1M NaCl solution at a rate of 0.13 ml/sec. Spreading of acidic area is observed in real time.

Fig.8 shows the experimental result of high-speed chemical imaging at 14 fps when 0.2 ml of 0.1M HCl solution was injected into 2.0 ml of neutral buffer solution at a rate of 0.13 ml/sec. After spreading of acidic area, shrinking of acidic area by buffering action is observed.

4.3. High-speed Imaging at 70 fps

Chemical imaging at even higher speed is possible by reducing the sampling number.

Fig.9 shows the experimental result of high-speed chemical imaging at 70 fps when 0.3 ml of 0.1M HCl solution was injected into 2.0 ml of 0.1M NaCl solution at a rate of 0.13 ml/sec. The quick spreading of acidic area is observed at time resolution of 14.3 msec.

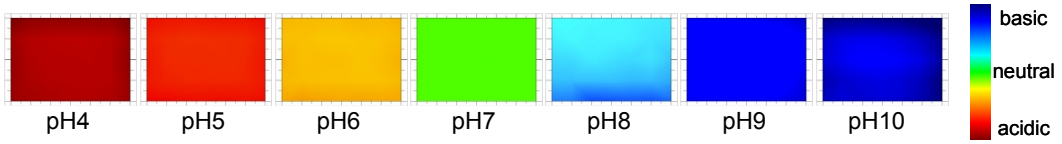

Fig.6 Chemical images of buffer solutions of pH4, 5, 6, 7, 8, 9, and 10.

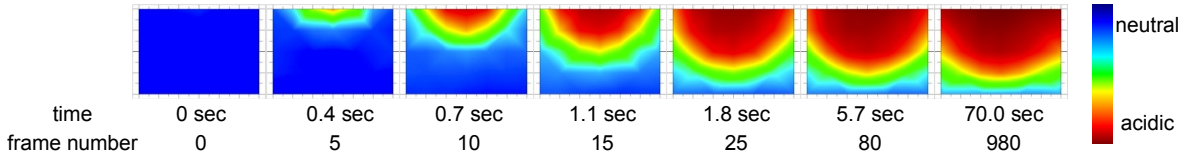

Fig.7 Example of high-speed chemical imaging at 14 fps (injection of HCl in NaCl solution).

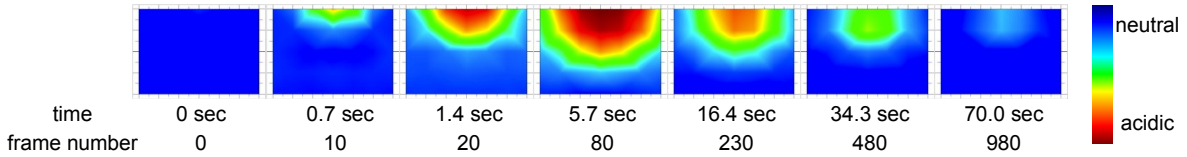

Fig.8 Example of high-speed chemical imaging at 14 fps (injection of HCl in buffer solution)

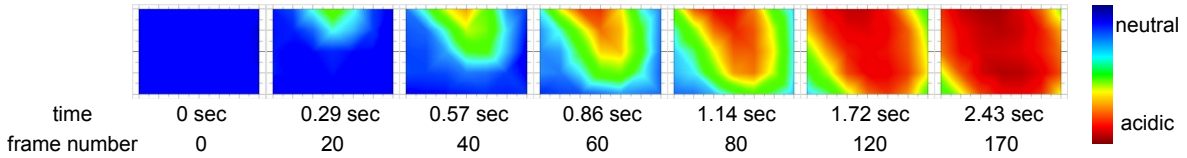

Fig.9 Example of high-speed chemical imaging at 70 fps (injection of HCl in NaCl solution)

5. Conclusion

In this study, a high-speed chemical imaging system was developed based on front-side-illuminated LAPS, for which higher modulation frequencies can be used in contrast to the conventional chemical imaging system with back-side illumination. The experimental results of chemical imaging demonstrated that chemical imaging by front-side illumination is also possible. Using the developed system, the quick change of pH distribution could be visualized as a sequence of chemical images acquired at 70 fps (time resolution: 14.3 msec) at a resolution of 5×7 pixels.

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