



High-speed chemical imaging system based on front-side-illuminated LAPS

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ABSTRACT

The chemical imaging sensor is a semiconductor-based chemical sensor that can visualize the spatial distribution of specific ions on the sensing surface. The conventional chemical imaging system based on the light-addressable potentiometric sensor (LAPS), however, required a long time to obtain a chemical image, due to the slow mechanical scan of a single light beam. 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 on the sensor plate by frequency division multiplex (FDM). However, the conventional measurement geometry of back-side illumination limited the bandwidth of the modulation frequency required for FDM measurement, because of the low-pass filtering characteristics of carrier diffusion in the Si substrate. In this study, a high-speed chemical imaging system based on front-side-illuminated LAPS was developed, which achieved high-speed spatiotemporal recording of pH change at a rate of 70 frames per second.

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1. Introduction

The light-addressable potentiometric sensor (LAPS) [1] belongs to the family of semiconductor-based chemical sensors with an electrolyte–insulator–semiconductor (EIS) structure. The width of the depletion layer in the Si substrate responds to the ion concentration on the sensing surface, and the variation of its capacitance is read out in the form of AC photocurrent induced by illumination of the sensor plate with a modulated light. The most advantageous point of the LAPS measurement is that the measurement area can be defined by illumination. Based on this addressability of LAPS, we developed the chemical imaging sensor [2], which could visualize the two-dimensional distribution of specific ions in the solution on the sensing surface.

In a conventional chemical imaging system based on LAPS, a scanning light beam was used to obtain a two-dimensional map of the ion concentration. Due to the mechanical scan, however, a typical scan time to acquire a chemical image at a resolution of 128×128 pixels was about 3 min, which was too long to analyze the dynamics of chemical reactions.

Frequency division multiplex (FDM) [3] is one of the methods for high-speed chemical imaging, in which a plurality of light beams modulated at different frequencies are employed to read out the ion concentrations at different locations in parallel as shown in Fig. 1. In this case, the photocurrent signal is a superposition of all frequency components and each component can be individually extracted by Fourier analysis. Based on FDM-LAPS, we developed a chemical image scanner system [4], which employed a linear array of 16 LEDs to scan a chemical image at a resolution of 16×128 pixels. The scan time was reduced to 6.4 s, which, however, was still slow in comparison, for example, to the pH imaging sensor based on the charge coupled device (CCD) technology [5,6]. In our previous study, we developed a chemical movie recording system based on FDM and a two-dimensional array of LEDs. Using a two-dimensional array of LEDs as a light source, the slow mechanical scan of the conventional system was replaced by much faster electrical switching and a high-speed imaging at a frame rate of 10 fps (frames per second) was achieved [7].

In FDM, the required bandwidth of the modulation frequency is given by (number of light beams) \times (frequency spacing) and the time window of Fourier analysis or the sampling time must be at least as long as $1/(\text{frequency spacing})$. For high-speed chemical imaging, therefore, a wide bandwidth of frequency is required to increase the multiplicity and to reduce the time window. Here, two different geometries are possible in LAPS measurement [8] as shown in Fig. 2. Those are front-side and back-side illuminations,

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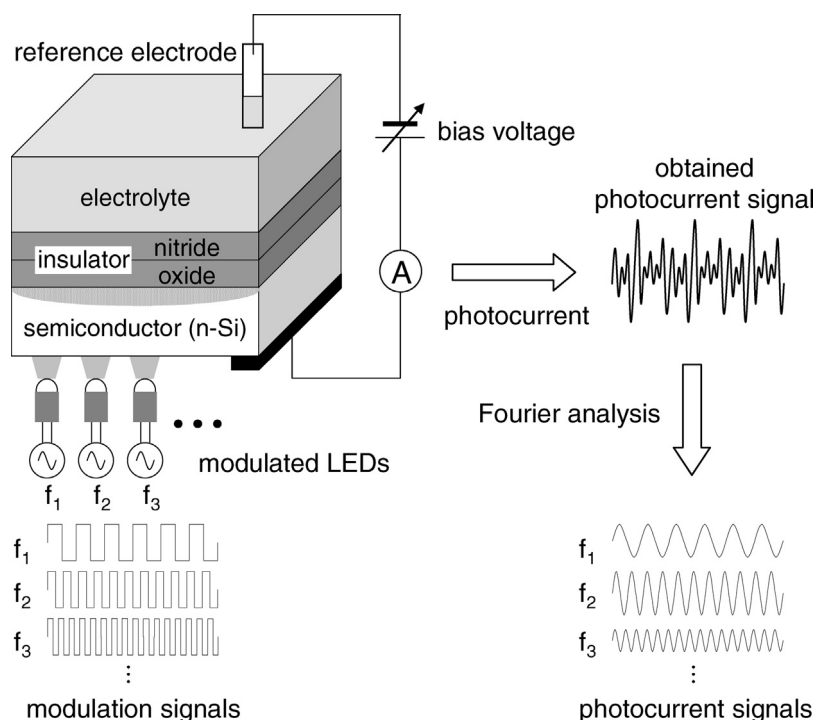


Fig. 1. Diagram of simultaneous measurement at multiple points on the sensing surface by frequency division multiplex.

in which the front-side or the back-side surface of the sensor plate is illuminated, respectively, by the modulated light. In the case of back-side illumination, which is the geometry of conventional LAPS measurement, the modulation frequency is limited by the low-pass filtering characteristics of carrier diffusion across the sensor plate. In front-side illumination, on the other hand, higher frequencies can be used, because photocarriers are directly generated and separated inside the depletion layer and no diffusion process is involved [8]. Front-side illumination, however, has some restrictions. For front-side illumination, the modulated light beam has to transit through the object to be measured. The object, therefore, must be light-transmissive and the distribution of the light transmittance of the object must be invariant while the pH distribution changes.

In this study, we developed a high-speed chemical imaging system based on the combination of FDM, a two-dimensional array of LEDs and front-side illumination. Front-side illumination allowed a wide bandwidth of the modulation frequency required for high-speed measurement by FDM. The developed system achieved high-speed chemical imaging at about 70 fps and provided real-time observation of a spatiotemporal change of pH distribution in a solution.

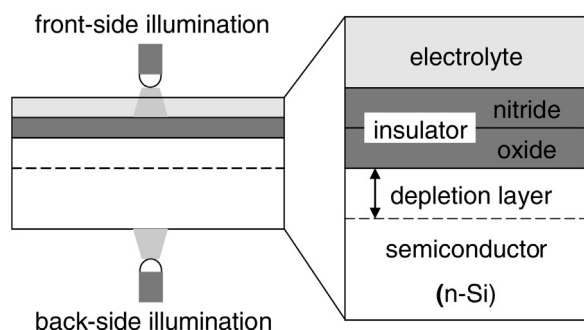


Fig. 2. Two different geometries of the LAPS measurement: front-side and back-side illuminations.

2. Experimental

Fig. 3(a) shows a schematic view of the high-speed chemical imaging system developed in this study. This system consists of a sensor plate, a two-dimensional array of LEDs (LED matrix), an oscillator array and a control PC with measurement software.

2.1. Sensor plate

The sensor plate (36 mm × 36 mm) used in this study was made of n-type Si wafer with a thickness of 200 μm and a resistivity of 10–20 Ωcm. The top surface of the sensor plate was covered with a 50 nm-thick thermal SiO₂ layer to obtain a semiconductor–insulator interface with low trap density and subsequently a 100 nm-thick Si₃N₄ layer deposited by low-pressure chemical vapor deposition (LP-CVD). The surface of the Si₃N₄ layer functions as a pH-sensitive surface when brought into contact with the sample solution. A thin ohmic electrode was deposited on the back-side of the sensor plate.

2.2. LED matrix

Fig. 3(b) shows the top view of the LED matrix with 7 × 5 LEDs (TA07-11SRWA, Kingbright Elec. Co., Ltd.) installed above the sensor plate as a light source. The peak wavelength and the intensity of illumination are 660 nm and 24 mcd, respectively. The light-emitting face of the LED matrix is immersed directly into the sample solution. The distance between the LED matrix and the sensor surface, or the thickness of the solution layer, through which the light has to pass, is about 0.6 mm. Five different frequencies are input to each column of the LED matrix and five points on the sensor surface corresponding to the five LEDs on one line are measured in parallel. By electrically switching the illuminating lines from the first to the seventh row in sequence, ion concentration values at all 35 points are obtained without mechanical scan.

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