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Time-resolved mapping of neurotransmitter fluctuations by arrays of nanocavity redox-cycling sensors

E. Kätelhön[a,b], B. Hofmann[a,b], M. Banzet[a,b], A. Offenhäusser[a,b], B. Wolfrum[a,b]*

[a]*IBN-2, Forschungszentrum Jülich GmbH, Jülich, Germany*
[b]*JARA- Fundamentals of Future Information Technology, Jülich, Germany*

Abstract

We introduce a novel device for the spatiotemporal detection of neurotransmitters on-chip. Our device features an array of individually addressable nanocavity sensors that utilize redox-cycling for the localized electrochemical determination of neurotransmitter concentrations. This effect is based on rapid, repeated redox-reactions between two closely spaced electrodes and results in a high amplification of the electrochemical signal of individual molecules. Each sensor is equipped with two independently biased electrodes that are separated by an ~65 nm wide cavity, thus enabling efficient redox-cycling amplification. The sensors feature entry ports in the micrometer regime and can be closely packed for the chemical mapping at high spatial resolutions.
In this paper, we present the devices capabilities in neurotransmitter detection. The sensors are electrochemically characterized and their functionality is demonstrated by measuring localized dopamine fluctuations in a microfluidic channel.

Keywords: Redox Cycling; Nanocavity; Dopamine; Electrochemical Sensing;

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1. Introduction

Numerous neurodegenerative diseases are caused by abnormal processes in neurotransmitter-based signaling. Hence, the investigation of inter-cellular communication via neurotransmitters plays a crucial role in current research. Various methods for the detection of neurotransmitters have been investigated during the last decades [1, 2, 3]. Particularly electrochemical methods have received distinct attention due to their capacity for simple online detection in situ. However, the mapping of single cell neurotransmitter release on a neural network scale still remains a challenge today. We follow a chip based approach for this application utilizing redox cycling amplification to increase the molecular sensitivity, see Fig. 1.

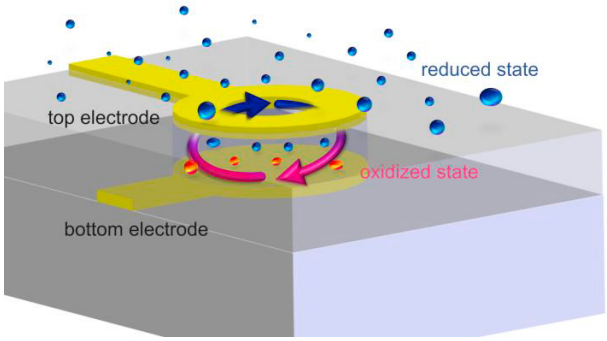

Fig. 1 Schematic drawing of the sensor layout and illustration of the redox cycling effect. Redox-active molecules in the reduced state are indicated by blue circles, molecules in the oxidized state are indicated by red circles.

2. Results

Our sensors were characterized using various concentrations of the redox-active neurotransmitter dopamine. According to the theoretical prediction, the experimentally obtained electrode currents were proportional to the applied dopamine concentrations. Thus, quantitative measurement of the dopamine concentrations were performed successfully. The sensor array was further investigated in a chemical gradient which was generated in a microfluidic channel (see Fig. 2). Throughout the experiment, a dopamine containing phosphate buffered saline (PBS) solution and a pure PBS solution were injected. At the two inlets, we changed the applied pressures periodically at a phase shift of about 180 degrees, while the chemical gradient in the channel was resolved at the sensor array. Figure 3 representatively shows the response of a single sensor. We observe periodical current traces, whose frequencies match the frequency of the applied pressures at the inlets. The sensor signals represent the local concentration fluctuations folded with the response function of the redox cycling sensor. Moreover, it can be seen that the anodic and cathodic sensor responses feature similar magnitudes, indicating a redox cycling efficiency close to 100%.

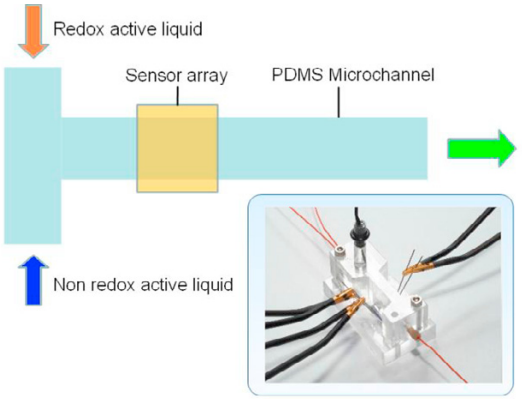

Fig. 2 Scheme of the electrochemical characterization of the sensor array in a microfluidic channel. Different chemical substances are injected via the two inlets, while the mixing gradient is detected by the sensor array. The photography in the bottom right corner shows the experimental setup.

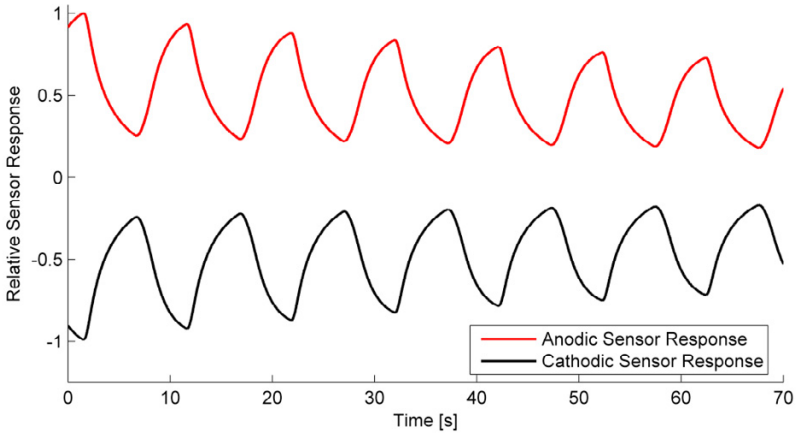

Fig. 3 Normalized response of a single sensor. The data was recorded using a 1mM dopamine solution in PBS and a pure PBS solution. The applied pressures were varied between 10 mBar and 100 mBar at a frequency of 0.1 Hz.

Acknowledgements

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