A NOVEL DYNAMIC ELECTROCHEMICAL TRANSDUCTION MECHANISM FOR LOW CONCENTRATION ANALYTE DETECTION


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ABSTRACT

In this work, we present a novel transduction mechanism by monitoring the dynamic charge decay of a capacitor used as the charge supplier for interdigitated array (IDA) microelectrode in redox cycle. The analyte concentration in the electrochemical cell has a decisive effect on the charge decay dynamics of capacitor, where lower concentration analyte causes slower charge decay. The transduction mechanism demonstrated in this work shows a potential for the detection of ultra-low concentration of analyte. A proof-of-concept of this novel transduction scheme has been successfully demonstrated by characterizing the dynamics of charge decay for a 1.0 μF capacitor, which is discharged by p-aminophenol (PAP) redox cycling at mM concentrations. Lower concentration detection towards pM range is currently under investigation.

KEYWORDS: Interdigitated array, dynamic transduction, low concentration, electrochemical detection

INTRODUCTION

For electrochemical detection, the interdigitated array (IDA) microelectrode has a good electrochemical self-amplification capability due to the redox cycling between two working electrodes, thus achieving high detection sensitivity. While IDA is used in many configuration modes [1, 2], it is typically used as an amperometric device. In order to detect very low concentration analyte, a three-dimensional structure that improves the redox species trap ratio [3, 4], a larger electrode surface area, or a narrow electrode space have been used to attain higher current signal. Alternately, a low noise electronic circuit can be used to measure very low currents from the IDA (e.g. current chopper amplifier [5]). The former needs a complicated fabrication process and the latter requires a complex circuit design.

In this work, a charge-coupled dynamic electrochemical transduction method for low concentration analyte detection is demonstrated to overcome these problems by changing the current output to a voltage output through a charged and noiseless capacitor as the source of charges for the electrochemical cell.
MECHANISM OF A DYNAMIC ELECTROCHEMICAL TRANSDUCTION

In the conventional configuration of IDA, a potentiostat provides charge to the electrochemical reaction on IDA by applying potential on the two working electrodes (WE1 and WE2). The redox cycling of analyte occurs on WE1 and WE2, and the currents from WE1 and WE2 are measured to analyze the redox species concentration. The novel electrochemical transduction mechanism uses a charged capacitor as the source to provide energy for the electrochemical reaction. The charges on the capacitor are consumed by the reaction in the electrochemical cell, which causes the capacitor voltage to decay. The analyte concentration in the electrochemical cell has a decisive effect on the capacitor voltage. For lower concentration analyte, the capacitor discharge is more gradual. Figure 1 illustrates that for a fixed amount of charge on the capacitor, the low concentration redox species on IDA consumes the charge slowly (Figure 1(a) and (c)), while the high concentration redox species consumes the charge quickly (Figure 1(b) and (d)). The charge consumption process is monitored by recording the dynamic voltage change across the capacitor.

![Figure 1. Illustration of the novel electrochemical transduction mechanism.](image)

EXPERIMENTAL SETUP

Figure 2 presents the measurement method for this novel mechanism, which consists of two operation phases. In phase 1, as shown in Figure 2(a), the switch is on, and the two working electrodes together with the capacitors are connected to two fixed potentials (V1 and V2), where V1 and V2 are the reduction and oxidation potentials, respectively. The IDA works in the conventional static mode and has two steady current outputs. The two capacitors are charged to V1 and V2. Capacitors and IDA-based electrochemical cell are independent components in this circuit. In phase 2, shown in Figure 2(b), the switch is off, and the charge on capacitor is used for further electrochemical reaction at the IDA.
The capacitor voltage decay is monitored by an instrumentation amplifier (IA) and used to calculate the analyte concentration.

Figure 2. Two step operations of the novel electrochemical transduction mechanism.

A microfabricated IDA with 80 pairs of fingers is used as the reaction electrode in the electrochemical cell, as shown in Figure 3. Each finger is 2 mm long, 10 μm wide with 10 μm spacing between neighboring fingers. The IDA (Gold), integrated with counter electrode (Gold) and reference electrode (Silver/Silver Chloride), is made by conventional microlithography techniques.

The measurements are done in N₂ environment in order to avoid the oxidation of PAP in air and a LabVIEW program controls the measurement circuit and samples data from the instrumentation amplifier (Figure 4).

Figure 3. Microfabricated IDA.

RESULTS

The IDA equivalent circuit has been modeled using SPICE. For the simulation, in phase 1 the IDA-based electrochemical cell is modeled as a current source and in phase 2 it is modeled as a voltage controlled current source. At the transition from phase 1 to phase 2, the equivalent current source (for the IDA) has current values in the range of 0.1 μA ~ 10.0 μA. This matches the IDA’s limiting current values corresponding to 0.1 mM ~ 10 mM analyte concentration.

The experimental results are shown in Figure 5(b). These capacitor voltage decay characteristics clearly show that lower concentration analyte causes slower capacitor
CONCLUSIONS

A novel dynamic electrochemical transduction mechanism for low concentration analyte detection has been developed in this work. Using this novel transduction configuration, the voltage decay characteristics on a 1.0 µF capacitor discharged by PAP redox cycling, at different concentrations, have been simulated and experimentally verified. We believe this novel electrochemical transduction mechanism has a great potential application in micro total analysis system (µTAS) for very low concentration analyte detection.

ACKNOWLEDGEMENTS

The authors thank Mr. Jeff Simkins, Mr. Rong Rong and Mr. Ron Flenniken in University of Cincinnati for their technical assistance in IDA microfabrication.

REFERENCE: