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Bipolar transistor amplifier for transduction of electrochemical response to visual perception

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ABSTRACT

A simple device for the transduction of an electrochemical signal to a visual readout suitable for point of care diagnostics has been designed. The transducer consists of an electrochemical cell and a 4-electronic components circuit, namely two resistors, one transistor and one light emitting diode (LED). The response from the electrochemical cell is amplified by the transistor providing output for the direct visual readout with the naked eye. Function of the device was verified in the experiments with hydrogen peroxide. Simple adjustment of the values of resistors provided tuneable sensitivity, positive threshold level and limit of detection as well. The biosensing abilities of the proposed system were tested on the analytical model using immobilised glucose oxidase. The principal benefits of the proposed platform include uniquely low construction costs and high simplicity. This approach is innovative in the transduction/conversion of the signal to visual perception and also in the signal generation – no potentiostat nor galvanostat are used compared to the majority of electrochemical measurements.

1. Introduction

Point-of-care (POC) diagnostics is an attractive analytical approach which is nowadays rapidly developing. Its merit stems from the transfer of an analytical act from a specialised laboratory close to the patient or generally directly to the site of sample collection. The production costs of analytical devices based on POC principles should be preferentially low resulting in low cost of one analysis. Furthermore, portability and simple operation are necessary and the obtained results should be suitable for straightforward interpretation even by unskilled person and without any operation manual [1,2].

A paper indicator still remains the simplest analytical POC system, e.g. litmus paper and urine test strips. Its main advantage originates from the construction material used paper. Cheapness, elasticity, temperature and dimensional stability, high liquid absorption, porous structure, capillary action, high ratio of active surface to volume, biocompatibility and simple degradability (either incineration or biodegradation) all belong to the favourable properties [2–4]. Devices based on paper substrate are highly suitable for disposable sensors; the low cost of one analysis is related to the possibility of mass production. The detection ranges are at milimolar concentrations for low molecular clinical analytes as glucose, lactate and uric acid [5,6]. Systems reaching submilimolar concentrations were reported for particular analytes, however, specialised instruments (readers) were necessary for the precise readout of the signal [7]. With some exceptions the obtained analytical information is principally in the YES/NO format.

Complex instruments which contain sophisticated electronics with potentiostat, microcomputer, display, software etc. [8] are on the opposite side of the spectrum of POC devices. These instruments exhibit high flexibility of measurement performance and complexity of the generated data. They are able to provide many measuring approaches, possibility to detect several analytes upon relatively undemanding modification of system, high sensitivity, the obtained value of concentration of an analyte is visualised as a number on the display and the possibility of local data storage and nowadays also the popular transfer to internet-based cloud systems is usually provided. The last option allows remote interpretation by expert and re-sending the decision back to “the sampling point”. The higher price and operating costs are common disadvantages of these instruments. The handling and operation need not to be necessarily simple. Moreover, with the increasing complexity of the instrument, the probability of a malfunction grows as well.

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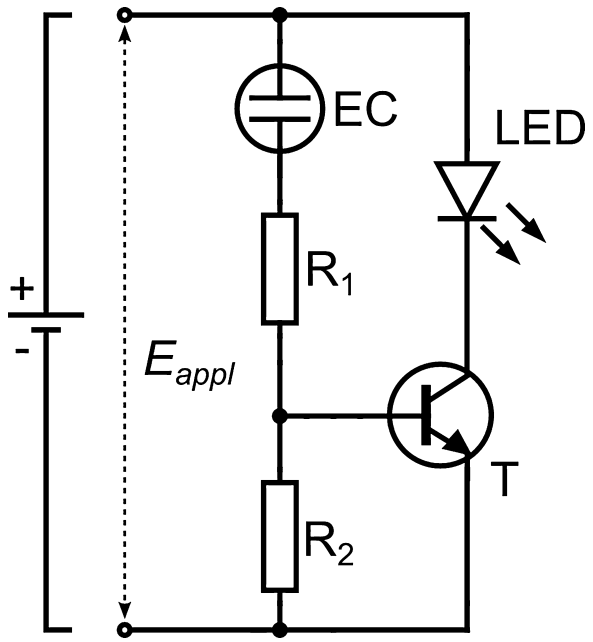
Generally, any diagnostic device can be described as a system consisting of sensing and reporting units. Sensing electrodes or generally analyte sensitive part represents the sensing unit and the reporting unit is typically some digital display. In the more sophisticated instruments, an additional unit responsible for amplification and conversion of chemical signal to analytical information is present; however, as this unit is not directly visible, it can be neglected or regarded as a part of the reporting unit. Sensing and reporting units are overlapped for paper indicators; the colour change of the analyte sensitive part (reporting unit) is usually due to the interaction of the analyte with other reagents (sensing unit) in the same place.

A diagnostic device as an interlink between the previously mentioned simple paper indicator and the sophisticated electronic instrument is still a challenging task. The diagnostic device combining simplicity and low costs of the paper indicator and sensitivity and wide applicability of electrochemical methods can be obtained by absolute simplification of the electronic circuitry of the common electrochemical transducers. Only fundamentally functioning electronic components will then be present omitting electronic display, microcontroller, sophisticated amplifiers etc. If the simplification of the reporting part is so drastic that it cannot proceed any further, the final stage of the most simplified reporting unit is only a single electronic component, e.g. light emitting diode (LED).

Signalling by LED is routine approach in electronics but it was rarely used in analytical applications. The signalling of an analytical event (detection) by light emitting diode has already been reported for two cases. First, presence of H_2S in the sample decreased electronic resistance of (poly)aniline layer thus modulating light emission of LED connected in series [9]. Secondly, LED connected in series with the auxiliary electrode in the common three electrode electrochemical set-up was used for reporting of redox reactions. Electrochemical conversion of iron and ruthenium salts was probed. The reporting of electrochemical reactions using LED exhibited better properties compared to the system based on electrochemical luminescence [10].

Both these concepts are limited by the threshold current upon which LED starts to emit light. This threshold current is in nowadays LEDs in the range of high microampers. This detection scheme is thus not suitable for applications where lower currents should be detected.

In our approach, the simplification of the reporting unit is only one step milder, the sensing unit is connected to the reporting unit through a simple amplifier. General electrochemical transduction platform is thus obtained by integration of a simple (e.g. bipolar) transistor amplifier with the electrochemical cell (consisting of anode and cathode) connected to its base electrode and light emitting diode behind this amplifier. As hydrogen peroxide is the most widespread tracer for enzymatic electrochemical biosensors based on oxidases, it was employed for characterisation of the performance of the proposed system. With the increasing concentration of H_2O_2 in the electrochemical cell, the resistance of the cell decreased resulting in opening of the transistor and consequent signalling by LED. The applicability of the proposed system was demonstrated on the well described model of electrochemical biosensing of glucose using glucose oxidase. Excluding power supply and electrochemical cell (sensing unit), a four component electronic circuit (two resistors, one transistor and one LED) represents promising electrochemical transducer which lowers the cost of the analytical device down to the level of paper indicators. With possibilities of an alternative power supply (e.g. solar cells and harvesters [11], a liquid sample as electrolyte of in-place-constructed batteries [12] and so called "fluidic batteries" [13]), the proposed system represents promising fundamental platform for POC diagnostics.



Scheme 1. Scheme of the simplest transduction of the electrochemical event using a bipolar transistor amplifier consisting of electrochemical cell (EC), resistors (R_1 and R_2), light emitting diode (LED) and transistor (T). E_{appl} denotes applied potential – a power supply.

2. Materials and methods

2.1. General setup

Measuring circuitry was constructed according to Scheme 1 using common electronic components from the local supplier (GM Electronics, Czech Republic). Electrochemical experiments were performed in the flow-through system, flow rate was 150 μl/min and buffer carrier was 50 mM phosphate, pH 7.4, containing 0.1 M KCl and 0.1% Tween 20. The measuring system was realised with the 4-channel screen-printed electrode (BVT Technologies, Czech Republic) inserted to the previously reported 4-channel flow through cell [14,15]. 4 platinum disc electrodes were connected in two pairs as anode and cathode. Total geometric area of each electrode was 1.57 mm^2. Changes of potential in the electrochemical system as a response to different concentrations of H_2O_2 (Penta, Czech Republic) were followed by Micro-Ohm Meter 34420A (Agilent) operated by in-house developed software LabTools. All other chemicals were of the highest purity available and used as received.

The voltage generated between anode and cathode of disconnected LED upon illumination with ordinary fluorescent bulb Osram Dulux 20 W/865 was measured by Handheld Digital Multimeter U1253A (Agilent).

2.2. Preparation of electrodes modified with glucose oxidase

4-channel screen printed electrode (SPE) was modified by the enzyme glucose oxidase (Sigma–Aldrich) according to the previously reported method [16]. Briefly, SPE was washed with acetone and distilled water. 1 μl of the enzymatic mixture consisting of bovine serum albumine 1.6 mg (Sigma–Aldrich), glucose oxidase 0.5 mg, phosphate buffer 163 μl and 2% glutardialdehyde 11 μl (Fluka) was deposited on the active area of all four electrodes. Thus modified electrodes were incubated in water saturated atmosphere overnight at 4 °C. After removal, electrodes were left to dry at laboratory atmosphere and were ready for use.