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Flexible ion sensors for bodily fluids

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Abstract

This work presents an array of ion selective electrodes combined with a solid state reference electrode for analyzing of potassium and sodium ions in fluid. The sensors was fabricated using screen printing and drop casting techniques. The ion selective electrodes were made from a stack of screen printed AgCl electrode and solid state polyhydroxyethylmethacrylate hydrogel internal electrolyte layers, which were preconditioned in 0.1 M KCl. The sensitivity of the sensors to potassium and sodium ions was achieved by casting a plasticized PVC based ion selective membrane, which contained an optimal ionophore to polymer ratio, directly on the hydrogel layer. The reference electrode was made on another AgCl electrode, which was covered with the same hydrogel. For both ions, sensitivities of 50 mV per decade were achieved in a concentration range 10^{-3} - 1 M. In this concentration range, the potassium sensor had no cross sensitivity to sodium ions and *vice versa*.

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1. Introduction

The aim of this work is to develop a flexible sensing platform, which allows to analyse multiple ions in fluids. Using low cost processes such as screen printing and drop casting on a large flexible substrate, the cost of miniaturized multiple ion sensors can be reduced. Earlier we have demonstrated a low cost patch consisting of chloride selective electrode and integrated reference electrode for real-time monitoring of chloride concentration in sweat [1]. To add more functionality to this platform, potassium and sodium selective electrodes were our next focus because these ions are important contributors in sweat and urine [2,3,4]. It has been shown that monitoring these ions is beneficial to control human physiological conditions. For example, an elevated concentration of sodium in sweat is reported as consequence of a high level of salt uptake and dehydration [3]. This low cost disposable sensing platform is especially

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suitable for quickly access the sodium and potassium levels in urine, which are found to be linked to disease such as hypertension [4,5].

In this work, the low cost potassium and sodium selective electrodes were developed on an array of the screen printed AgCl electrodes with a solid state internal electrolyte layer (see Fig. 1). The sensitivity of the sensors to potassium and sodium ions was achieved by casting a plasticized polyvinylchloride (PVC) based ion selective membrane containing an optimal ionophore to polymer ratio, directly on the hydrogel layer. The sensitivity, selectivity and detection limit of the sensors were characterized. The influence of the ionophore concentration on the sensitivity and detection limit of the sensors was part of this study as well.

2. Experiments

The potassium and sodium sensors were developed on the AgCl electrodes, which was fabricated by screen printing AgCl conducting paste (Dupont 5876) on a PET foil. After the printing, the AgCl layer were dried at 110°C for three minutes and a next AgCl layer was printed to achieve the required thickness. The isolation layers were printed using DuPont 8153 insulating paste on the dry AgCl electrodes using the second mask. The printing condition and thickness of the AgCl and isolation layers were optimized in a previous study to achieve the optimal performance of the AgCl electrode [1].

To form the internal hydrogel electrolyte for the ion selective electrode, an O-ring with an inner diameter of 3 mm was glued on the AgCl electrode with 3M Scotch Weld Epoxy (DP460) and cured at room temperature. The internal gel-like electrolyte layer was made from a mixture of UV-sensitive hydroxyethylmethacrylate (HEMA) and a photo initiator and drop casted on the AgCl electrode inside the O-ring [1]. After curing under an UV lamp, the hydrogel layer was conditioned in 0.1 M KCl solution for one day. The ion selective membrane was applied on the pHEMA surface inside the O-ring by drop casting of ion selective membrane solution containing polyvinylchloride, plasticizer, potassium tetrakis(4-chlorophenyl)borate, cyclohexanone and corresponding ionophore (2.7 wt % valinomycin for K⁺ and 1.1 wt% sodium ionophore IV for Na⁺) as seen in Fig. 1. After drying overnight, the ion selective electrodes were ready to test. The reference electrode was made on another AgCl electrode, which was covered with the same hydrogel.

Calibration of the potassium and sodium selective electrodes was performed in a series of test solutions containing 10⁻⁴ - 1 M of the ion of interest and 0.1 M of another cation as a supporting electrolyte, both with chloride as anion. All chemicals used were of analytical grade.

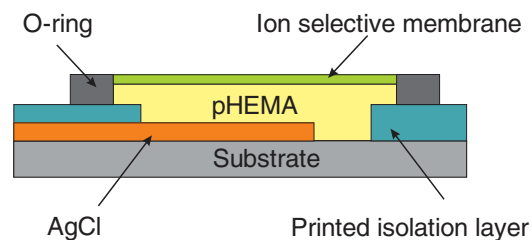


Fig. 1. Illustration of cross section of an ion selective electrode with a hydrogel pHEMA layer casted within an O-ring, which was glued on top of the AgCl electrode. An ion selective membrane was casted on top of the pHEMA layer, which also covered part of the O-ring.

3. Results and discussions

The sensitivity of a potassium selective electrode was determined by measuring its potential *versus* a 3 M KCl commercial reference electrode (CRISON 5044) in solutions containing 10⁻⁴ – 1 M KCl and 0.1 M NaCl. The potential – time trace of the potassium selective electrode with increasing and decreasing KCl levels is presented in Fig. 2a. This figure shows that the potential sharply increased with increasing potassium concentration. When the potassium concentration was reduced from 1 M to 10⁻⁴ M, the potential changed back to the same potential level, as was observed during the first exposure to 10⁻⁴ M KCl showing good reproducibility of the sensor. The equilibrium potential of the potassium selective electrode in different KCl solutions extracted from the potential plateau levels in Fig. 2a is plotted

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