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A flow-through potentiometric sensor for an integrated microdialysis system

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Summary: In this paper, the performance of a flow-through potentiometric sensor based on a semi-permeable dialysis tubing implemented in silicon, is presented. The sensor is designed as part of a lab-on-a-chip system and has been successfully incorporated into an integrated microdialysis system. Results of a potassium sensor, operating stand-alone, and as part of the integrated system are presented.

Keywords: potentiometric sensor, flow-through system, potassium sensor, microdialysis, Lab-on-a-chip, μ TAS,

Category: 5 (Chemical sensors)

1 Introduction

A generic flow-trough potentiometric microsensor based on a semi-permeable tubing made in Perspex[®] has been described previously by Böhm [1]. As proposed in this paper the microchannel itself is an integral part of the sensor geometry and is formed by a tubular semi-permeable membrane. The major advantage of this geometry is the fact that commercially available ion-selective cocktails can be applied, yielding sensors for different analytes. The sensor based on this concept is designed in the form of an integral component of lab-on-a-chip systems.

The sensor fabrication and the performance of the potassium microsensor operating stand-alone, and as part of an integrated microdialysis system are presented.

2 Experimental

2.1 Materials

The dialysis tube from regenerated cellulose with MW cut-off of 20 kDaltons was adapted from an artificial kidney Filtral[®] 6, AN69 HF, Hospal France. Epoxy resin (Hysol[®], Dexter, USA) was used to fix the dialysis tube. The sensors windows were closed with a piece of Pyrex fixed with UV curable glue (Loctite 350).

To form the electrodes, a conducting silver-silver chloride paste (Electrodag[®] 6088 SS, Acheson Colloiden B.V., The Netherlands) was used.

Components of hydrogel: acrylamide AAm, N,N-bisacrylamide (crosslinker) 2,2-dimethoxy-2-phenylacetophenone DMPAP (photoinitiator) obtained from Fluka, and DMSO.

2.2 Sensor fabrication

Originally, the sensors were designed to be filled with two liquids: internal electrolyte and ionophore

cocktail. However, it was found that due to specific configuration of the sensor, it was difficult to form well defined interface between the two liquids. To solve the problem a UV sensitive hydrogel based on polyacrylamide was applied for entrapment of the internal electrolyte.

A cross section of the potentiometric flow-trough microsensor is shown in Fig. 1.

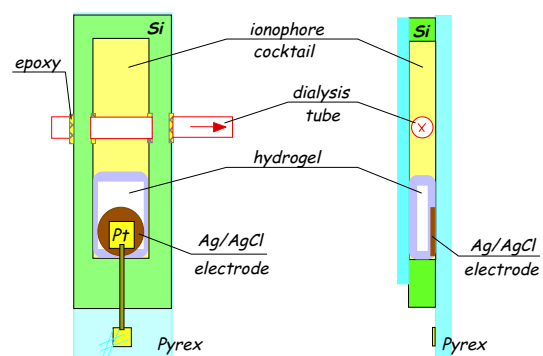


Fig. 1. Layout of the flow-trough potentiometric microsensor implemented in silicon.

The estimated swelling capability of the hydrogel containing 3.6% of a crosslinker vs. AAm was about 45%. The photographs in Fig. 2 show the pAAm hydrogel after drying at room temperature for 30 min and in a swollen state.

Before use the hydrogel was soaked in the internal electrolyte for at least 24 hrs. Prior to placing the hydrogel into the sensor cavity, it was cut to the proper square size. Then the hydrogel was placed in the sensor cavity and dried for about 15 to 30 min. Afterwards the cavity was closed with a piece of Pyrex and fixed with the UV curable adhesive.

Next the cavity was filled with the internal electrolyte to allow the hydrogel swell again for at least 1 h. Finally the excess of internal electrolyte in the surrounding of the dialysis tube was removed

and replaced with an ionophore (valinomycin) cocktail (60031 obtained from Fluka).

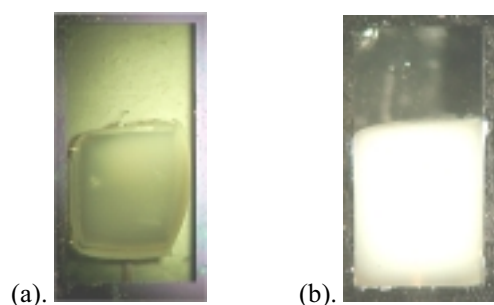


Fig. 2. The pAAm hydrogel in a dry state (a), and in a swollen state (b).

To form a reference electrode, the sensor cavity located next to the ion-selective one was completely filled with pAAm hydrogel saturated with 3 M KCl.

2.3 Measurements

The stand-alone sensors were operating at a constant continuous flow rate of 2 ml/h. The concentration of the potassium ions was changed by a standard addition method using 0.1 M and 1 M KCl aqueous solutions. As a background electrolyte 100 mM NaCl was used.

Sampling was performed using the integrated microdialysis probe under continuous flow at a flow rate of 2 μ l/min for 60 sec.

The potential of the detecting electrode was measured against the integrated reference electrode, using a home-made high-impedance instrumentation amplifier. Before measurements the sensors were allowed to stabilize by flowing with 50 mM KCl solution for about 2 hrs.

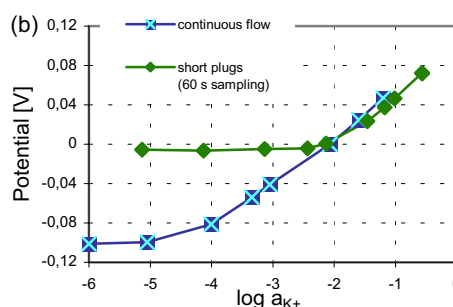
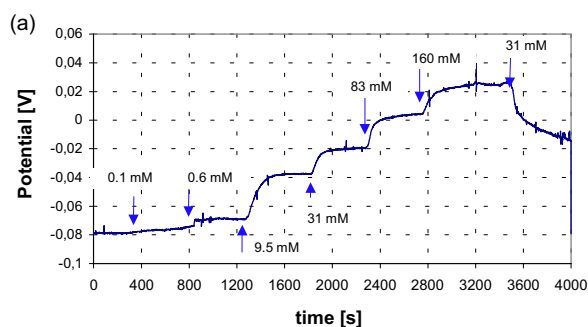


Fig. 3. Time response (a) and calibration curve (b) for the potassium sensor for continuous flow. Background electrolyte was 100 mM NaCl. In the upper curve of fig. (b) also the calibration curve for the sensor as integrated in the microdialysis system is shown.

References

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3 Results and discussion

The results presenting the dynamic response of the sensor and calibration curves for the continuous flow at a constant flow rate of 2 ml/h are shown in Fig. 3 a and b respectively. The sensitivity of the sensors varied from 50 to 55 mV/dec. The sensitivity depends on the time of the sensor conditioning (stabilization) before measurements. The minimal time for the sensor conditioning is 2 hrs.

The activities of the cations in the aqueous solution were calculated according to the Debye-Huckel approximation [2]. The potentiometric selectivity coefficient for the sensor for potassium over sodium ($K^{pot}=161.5$) was estimated according to the fixed interference method (FIM) [3] by increasing the concentration of the activity of the primary ion in the solution in steps of about 0.7 decade.

The sensor has been successfully integrated in a microdialysis system [4]. A calibration curve for the sensor operating in the microdialysis system for 60-second sampling through a microdialysis probe at a flow rate 2 μ l/min is shown in Fig 3.b (upper curve).

4 Conclusions

A flow-trough potassium potentiometric microsensor based on semi-permeable tubing has been implemented in silicon, and exhibit satisfactory parameters: near Nernstian response and a good selectivity over sodium.

The sensors have been successfully integrated into a chip resulting in lab-on-a-chip type device for microdialysis, consisting of several functional blocks: microdialysis probe and sensor array.

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