Journal of Diabetes Science and Technology
Volume 3, Issue 4, July 2009

ORIGINAL ARTICLES

Enhanced Glucose Sensor Linearity Using Poly(Vinyl Alcohol) Hydrogels

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Abstract

Background:
High linearities, sensitivities, and low oxygen dependence constitute prime requisites for electrochemical glucose sensors. However, for implantable sensors the need to control tissue inflammation requires the use of outer membranes that permit inward analyte diffusion while continuously releasing anti-inflammatory drugs and other tissue response-modifying (TRM) agents. We have shown previously that while outer membranes based on layer-by-layer (LBL) assembly enhance linearity, poly(vinyl alcohol)(PVA) hydrogels loaded with TRM-containing microspheres enable a significant reduction in tissue inflammation. This article discusses the amperometric performance of glucose sensors coated with stacked LBL/PVA hydrogel outer membranes.

Methods:
Sensors were fabricated by immobilizing glucose oxidase enzyme on a 50-μm platinum wire followed by deposition of stacked LBL/PVA hydrogel outer membranes. The sensor response to various glucose concentrations was determined by applying 0.7 V vs an Ag/AgCl reference electrode in phosphate-buffered saline (37°C). Michaelis–Menten analysis was performed to quantify sensor performance in terms of linearity ($K^{app}_{m,glu}$) and oxygen dependence (K^{app}_{m,O_2}/[Glucose]).

Results:
When overlaid onto LBL-assembled outer membranes, PVA hydrogels improved sensor linearity by 60% from 10 to 16 m*M* of glucose and resulted in a twofold decrease in oxygen dependence.

Conclusions:
Enhancement in the performance of a PVA-coated sensor is attributed to the oxygen-storing capability of PVA hydrogel due to the formation of hydrophobic domains during its freezing and thawing employed to physical cross-link the PVA. Such membranes with the capability to release TRMs continuously while storing oxygen constitute a major improvement over current outer membrane technologies.

J Diabetes Sci Technol 2009;3(4):863-874

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Abbreviations: (FAD) flavin adenine dinucleotide, (FT) freeze–thaw, (GO_x) glucose oxidase, (HA) humic acid, (LBL) layer by layer, (LRS) linear range sensitivity, (PBS) phosphate-buffered saline, (PPD) *ortho*-phenylenediamine, (PSS) poly(sodium 4-styrenesulfonate), (Pt) platinum, (PVA) poly(vinyl alcohol)

Keywords: apparent Michaelis-Menten constants, biosensor, freeze-thaw cycle, layer-by-layer assembly, linearity, outer membranes, oxygen content, oxygen dependence of biosensors, PVA hydrogels

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