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Uniform sensing layer of immiscible enzyme-mediator compounds developed via a spray aerosol mixing technique towards low cost minimally invasive microneedle continuous glucose monitoring devices

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Abstract

In this study, a uniformly mixed sensing layer of typically immiscible compounds, such as tetrathiafulvalene (TTF) mediator and glucose oxidase (GOx) enzyme, was developed using a simultaneous spray deposition technique ideal for mass production of glucose sensors at low cost while exhibiting enhanced amperometric response. For comparison, the sensors were fabricated via three different methods: conventional drop-cast of TTF and GOx compounds in subsequent layers (DL), spray deposition of the compounds in subsequent layers (SL), and spray mixing of the compounds as one uniform layer (SM). Uniformity of the sensing layers was investigated via Scanning Electron Microscopy (SEM) and Energy Dispersive X-Ray Spectroscopy (EDX) techniques demonstrating an even distribution of the TTF and GOx throughout the sensing layer for the SM sensors. The amperometric studies showed a significantly larger maximum current response, I_{\max} and sensitivity for the SM sensors as compared to the SL and DL sensors. The significantly better performance of the SM sensors correlated well with the even distribution of TTF and GOx throughout the sensing layer, resulting in enhanced electron transfer and redox reaction between GOx and TTF. The SM spray technique was then applied to deposit a uniformly mixed sensing layer on to 3D microneedle arrays to provide minimally invasive continuous glucose monitoring (CGM). In-vivo studies showed amperometric response from the microneedle CGM device was compatible to changes in blood glucose levels measured via the standard finger prick tests. Importantly, the deposition technique is suitable for mass production of the microneedle CGM at very low cost.

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