Wearable monitoring of wound pH through a flexible, fibrebased pH sensor

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Abstract

There is an increasing demand for technologies that can monitor the pH of wounds and of skin in real-time, which indicates the progress of healing and the physiological condition of the skin. We have developed a novel wearable platform for the real-time monitoring of the pH of skin and wounds. A highly conductive flexible cotton sensor fibre was fabricated. This exhibited significant antibacterial properties and a rapid, selective Nernstian sensitivity (vs. a wearable fabricated Ag/AgCl pseudoreference electrode) over the broad pH range of 2.0-12.0 when analysing only a $50~\mu$ l sample of pH-adjusted artificial sweat matrices.

Key words: Wearable sensor, conductive cotton, fibre electrode, pH, solid state

Introduction

Skin diseases, like eczema and psoriasis, are a common burden in the UK, especially in children, where 34 % suffer from such diseases at some point.1 Wound care and management is also a significant burden to the UK healthcare system, estimated at an annual cost of £5.3 billion, partly due to inappropriate diagnosis.2 Epidermal pH gives an indication of the physiological condition of the skin and the healing progress of wounds.3 An effective pH-sensing dermal patch would provide non-invasive skin and monitoring, aiding treatment. The aim of this work is to develop a fibre-based flexible electrode to measure skin/wound pH. This will facilitate point of care analysis and allow appropriate care to be administrated by medical professionals.

Methods

Conductive fibre electrodes were made by coating cotton with a conductive dispersion. The conductive dispersion used contained 0.5 wt.% polyethylenedioxythiophene (PEDOT) and 1.5 wt.% multi-walled carbon nanotubes (MWCNT). pH-sensitive Wearable electrodes fabricated by coating the conductive cotton with polyaniline (PANi) via potentiostatic electropolymerisation. PANi is a pH-sensitive polymer. Wearable reference conducting electrodes were fabricated by coating gauze with an Ag/AgCl ink. The subsequent (potentiometric) pH sensitivity of the PANi-coated fibres electrodes was measured against gauze-based reference electrodes.

For the antibacterial test, solutions of wild type *Escherichia coli* with yellow fluorescent protein were added to bare and conductive cotton yarn and incubated at 37 °C for 4 h. Each "biofouled" fibre was subsequently analysed using fluorescence microscopy at 488 nm excitation. The mean intensity of fluorescence was measured across a fixed area (0.217 mm²).

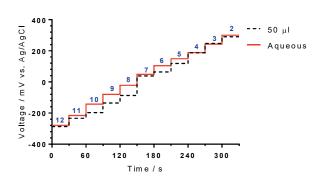
Results

The fibre-based pH sensor was found to maintain the flexibility of the cotton substrate, making it ideal for wearable applications. The sensor response was measured against a gauze-based reference electrode giving a Nernstian response of 59 ± 2 mV pH⁻¹ obtained over the pH range 2.0 to 12.0. This Nernstian response was also maintained as the volume of analyte was reduced down to 50 µl both in buffered solutions (58.9 ± 1 mV pH⁻¹) and in pHadjusted artificial sweat matrices (57 ± 2 mV pH-1) (Fig. 1(a) and (c)). A fast response time of 60 s (or less) was obtained (Fig. 1(a)). Furthermore, addition of interfering ions that are commonly found in sweat led to negligible voltage changes compared to the voltage changes due to pH variations (Fig. 1(b)).

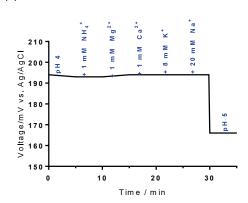
The successful operation over the pH range 4.0 - 9.0 is essential for the application in a wound

pH sensor because chronic wounds have a maximum pH of 9.0 and healthy, unwounded skin has a pH of 4.0 (Fig. 1(c)).^{4,5} Additionally, for the use in an epidermal patch, this sensor could monitor the critical skin pH range pH 4.0 - 7.0.⁶

(a)



(b)



(c)

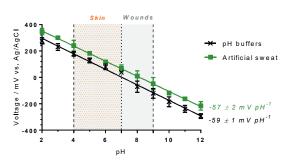


Fig. 1 General response of solid state pH analysis: (a) Response in 20 ml and in 50 μ l solutions. (b)

Selectivity when various sweat-related interferences are added to the pH 4 aqueous samples (20 ml). (c) Response in pH buffers and in pH-adjusted artificial sweat matrix (50 μ l). The error bars represent the standard deviation of three fibre repeats.

Bacterial colonization can delay the healing of chronic wounds therefore the antibacterial properties of these materials are also of interest (Fig. 2). Compared to bare cotton, the conductive cotton yarn showed significant antibacterial properties when incubated in the presence of fluorescent *E. coli*; a notably lower fluorescence intensity, hence lower bacterial growth on the surface, was observed. This may be explained by either the toxicity of the conductive cotton towards the bacteria or prevention of bacterial adherence. Further studies are underway to fully characterize these effects.

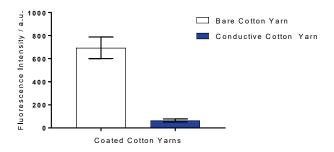


Fig. 2. Intensity of fluorescence (FITC, 488 nm) across a fixed area (0.217 mm2). Error bars represent standard error of three independent results.

Conclusions

The developed flexible pH electrode has a Nernstian response (vs. fabricated reference electrode) across a wide operational pH range (2.0 — 12.0) with minimal responses to interferences present in sweat. This intrinsically antimicrobial sensor is targeted at the real-time, non-invasive monitoring of wounds in healthcare. The conductive cotton fiber concept provides further opportunities for use in other real-time, on-body, flexible sensors for a variety of fields (advanced biomedical devices, smart electronics and wearable sensing products).

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