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Water-proof Anti-drying Enzymatic O_2 Cathode for Bioelectric Skin Patch

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Abstract

Herein, a water-proof anti-drying cathode for deformable bioelectric skin patch was developed by laminating a thin polydimethylsiloxane (PDMS) membrane (under 50 µm). The bilirubin oxidase (BOD) was modified on the electrode made of carbon fabric (CF) and employed as an air-breathing cathode. Adhered the PDMS membrane and the CF electrode properly, microspace serving as the air tank was formed at their interfaces. Those space enabled the operation of the patch even in a soaked condition for more than a few min without losing enzyme activity. Also, the anti-drying effect of PDMS prolonged the lifetime of the device wrapped to a finger for more than 3 hours with keeping transdermal current of ~ 20 µA cm^{-2}.

1. Introduction

Electrical skin patches for generating transdermal electrical current have been widely studied for anti-perspiration, drug delivery and tissue fluid collection. We have previously developed an electrical flexible skin patch with enzymatic biobatteries[1)] (Fig. 1). In this study, a water-proof breathable enzymatic electrode was developed, and its stable performances as the cathode of a bioelectric skin patch were demonstrated[2)].

A thin membrane of PDMS, which had high oxygen permeability, was used as the cover of the cathode electrode to maintain the three-phase boundary longer. Also, the PDMS membrane and the BOD-modified CF were adhered so that a microspace serving as the oxygen tank was formed at their interface, resulting in operation even in a soaked condition.

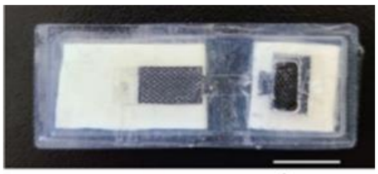

Fig 1. Photographs of skin patch.

2. Theory and Experiment

Enzymatic biobatteries generate electricity via enzymatic oxidation of fuels such as sugars in anode and reduction of ambient oxygen in cathode. They enable construction of the metal-free, disposable electrical skin patch. The reduction reaction in the air-breathing cathode is actively occurred at the three-phase boundary (solid; enzyme, liquid; electrolyte, gas; oxygen in the air). The performance of a cathode depends on the condition of the three-phase boundary so that the stabilization of the interfaces is essential to maintain patch performance. To cover the interface of the cathode, the PDMS precursor was spin-coated and gently coated CF with an uncured PDMS. The BOD cathode with modified CF was fabricated and we observed the reduction current while the electrode was placed in water and the relationship between the evaporation of the electrolyte from the patch and the current.

3. Results and discussion

3.1. Water-proof performance of PDMS membrane-integrated enzyme cathode

Fig. 2 shows the time-course of the electrode potential upon soaking, measured at a constant current of 0.5 mA cm^{-2}. In case of the without a PDMS membrane, the electrode potential immediately decreased due to the collapse of the three-phase boundary condition. On the other hand, the PDMS/CF modified BOD-cathode maintained its potential for 200 s, owing to the microspace formed due to the CF structures would serve as the air tank to ensure the observed longer-term operation underwater condition.

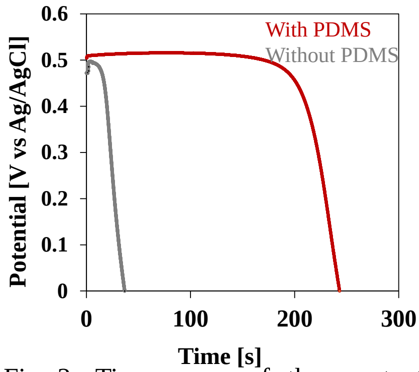

Fig 2. Time-course of the potential soaking at a constant current.

3.2. Anti-drying performance of PDMS membrane-integrated enzyme cathode

The bioelectric skin patch is well-wrapped around the index finger, indicating that the PDMS-attached electrodes cause no effect on the flexibility of the battery (Fig. 3a). In Fig. 3b, the stability in the transdermal current generation is compared between patches with and without the integration of PDMS film. At the initial stage, similar current around 25 μA cm^{-2} was recorded for both patches. In case of the patch without a PDMS membrane at cathode, the current decreased suddenly after 90 min. It was also visually conformed after the measurements that the patches without PDMS were dried by moisture evaporation. On the other hand, the patch with the PDMS membrane maintained the current value even after 180 min. The prolonged lifetime of the PDMS/CF-based cathode would be owing to the anti-drying effect of the PDMS membrane even when the patch is wrapped to a curved surface such as a finger.

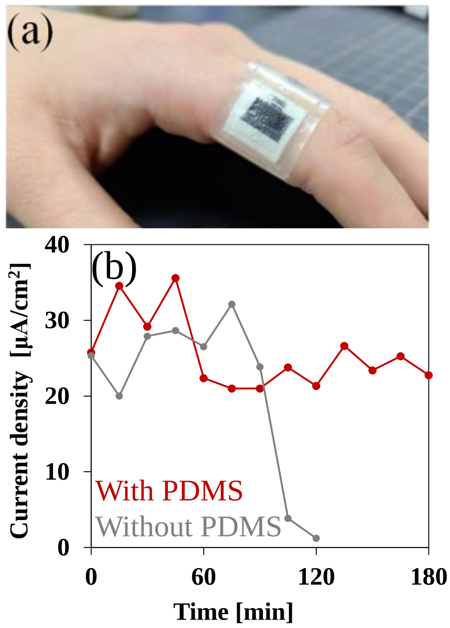

Fig 3 (a) Image of the bioelectric skin patch wrapped around a finger. (b) the transdermal current on the skin patches.

4. Conclusions

The integration of a thin PDMS membrane showed multiple effects in improving the practicality of the bioelectric skin patch with enzymatic O_2 cathode. First, the anti-drying effect enabled the operation time of the patch for more than 3 hours on skin at 37 °C. Also, the microspace at the interface between the PDMS and CF served as an O_2 tank that enabled the use of the patch even in wet conditions such as during the daily hand wash and taking a bath.

Reference

(1) S. Yoshida et al., JPhys Energy (2020) 2, 044004
(2) D. Terutsuki et al., J. of Power Sources (2022) 546, 231945.