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Abstract

Improvement in Limit of Detection of Paper-Based Electrochemical Enzymatic Biogas Sensor †

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Breath analysis is an attractive method for chronic diseases because it offers noninvasive, point-of-care diagnostics. Biomarkers of chronic diseases and many volatile compounds are included at low concentrations (sub ppm to ppt levels) in human breath [1,2]. Thus, gas sensors for exhaled breath require high selectivity and low limit of detection (LOD).

A study of highly selective biogas sensors utilizing the specificity of enzymes has been reported [3]. The detection area is fabricated simply by using chromatography paper where the enzyme/mediator solution is dried and immobilized. The $[Fe(CN)_6]^{4-}$ (Ferro), one of the mediators, changes to $[Fe(CN)_6]^{3-}$ (Ferri) due to the enzyme reaction when the target gas is blown on the detection area. Then the Ferri is reduced by the electrode with negative potential, and the current flows. However, it is difficult to measure low concentration substances in exhaled breath using the previous gas sensor because of the background current (BC: current at zero concentration), and its standard deviation is large. We assumed that the BC is caused by degradation of Ferro into Ferri during drying process because of the redox potential relationship between dissolved oxygen and Ferro.

This study is an attempt to decrease the background current and lower the limit of detection. We measured BC by optimizing the conditions of mediator, dissolved oxygen, and Ferro concentration, as well as temperature and time in drying process. The optimum condition was obtained with removing dissolved oxygen as soon as possible, Ferro concentration 3 mM, drying temperature 40 °C, and drying time 20 min. In this condition, LOD of ethanol gas sensor was improved from 39 ppm to 15 ppm (61.5%).

This result presents a new possibility which can be applied to biosensors for low concentration substances in human breath and diagnosing chronic diseases.

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