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Durable Textile-based Sensor for Repetitive Glucose Measurement

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Diabetes is one of a disease with high growth in this world. At the end of 2030, the World Health Organization predicts that the number of cases of diabetes worldwide will exceed 500 million, considering this condition preventive and corrective action are needed through simultaneous measurement of blood sugar levels. A new method has been developed that allows the blood sugar checking levels to be carried out continuously to achieve a comprehensive assessment non-invasive. This research studied the process of designing and fabricating a single-layer textile-based sensor through the direct coating method. The sensor's capabilities were assessed using a glucose solution with concentrations ranging from 10 to 50 mM at a voltage of 0.55 V for 300 seconds with a time interval of 0.5 seconds. Subsequently, durability testing of the sensor against repeated folding was conducted by subjecting it to repeated 90° folds with intensity variations of 0 to 50 times before chronoamperometry measurements were executed. It is observed that the design and fabrication of conventional single-layer textile-based sensors can be achieved using the coating method with carbon paste and Ag/AgCl paste on double-faced fabric. The single-layer textile-based sensor in this research demonstrates superior durability compared to screen-printed carbon electrode sensors.

Keywords: Biosensor, Diabetes, Textile-based sensor, Multi-layer, Non-invasive biosensor

1. Introduction

Health measurement instruments, encompassing those designed for the monitoring of cholesterol, lactic acid, and glucose, are expected to manifest exceptional operational efficacy over their lifespan [1]. The functionality of the sensor is heavily dependent on the electrodes integrated into these instruments. The size of the working electrode's surface is a crucial factor influencing overall performance. This surface is coated with an enzyme that acts as a catalyst, accelerating the oxidation process. This process involves the transfer of electrons, ultimately generating an electrical current [2]. Increasing the size of the working electrode's surface provides more space for the oxidation reaction to occur. This speeds up the reaction and produces a stronger electrical signal. A heightened electrical signal has the potential to augment measurement sensitivity and precision, ultimately culminating in enhanced sensor performance [3]. Nevertheless, limitations exist regarding the dimensions of the sensor, as it is preferable to maintain its compactness. One innovative solution to this problem is to use a textile-based sensor as the foundation material. Textiles have a porous structure that allows fluids to pass through easily from one side to the other. This is a major advantage for sensors as it means the substance being analyzed can be applied to both sides of the fabric.

Textile-based sensors created by directly applying materials to the fabric are ideal due to their flexibility, portability, non-invasiveness, and lightweight nature. Their effectiveness depends heavily on the careful design of the fabric structure, including how layers are arranged and electrodes are positioned. It's essential to place the electrodes precisely to prevent them from touching before the sample is added, which could cause a short circuit. The sensor should also allow liquid samples to easily spread across the entire surface of the electrodes. This ensures that the smallest amount of sample is needed to start the chemical reactions and detect the substance quickly. Therefore, a well-designed textile sensor is crucial for achieving optimal performance.

Double-layered woven fabrics offer a significant advantage in preventing short circuits during the electrode coating process due to their thicker construction. The fabric's unique structure, with two distinct layers of warp threads, creates a larger gap between the top and bottom surfaces compared to standard fabrics. This layered design acts as a moisture barrier, effectively controlling the movement of liquids and vapors within the fabric [4,5]. It's expected that this fabric design will effectively keep the electrode material on one side of the fabric during the manufacturing process [6]. The fabric's structure is influenced by several factors, including how tightly the weft threads are packed together, which affects the fabric's overall density. [7–10]. The objective of this research is to fabricate a textile-based sensor employing a single-layer concept through the direct coating method, utilizing carbon ink-coated yarn as the raw material for the working electrode and counter electrode. Additionally, Ag/AgCl ink-coated yarn is employed as the raw material for the reference electrode. The sensor's performance is assessed through measurements involving glucose solutions of varying concentrations. Furthermore, the durability of the sensor is characterized concerning repeated folds and its resilience to bending at various angles.

2. Methods and Methodology

(a) Chemicals and Materials

Silver/silver chloride and carbon pastes were procured from Kayaku Advance Materials (US). Potassium chloride was obtained from Merck (US), and Potassium Ferrycyanide was sourced from Sigma Aldrich (Germany). Phosphate buffer saline with a pH of 7.4 was acquired from Biogear (Indonesia), and Aqua bidest was purchased from Ikapharmindo Putramas (Indonesia). The warp yarn utilized was a blend of polyester-viscose (65:35) sourced from Dhanar Mas Concern (Indonesia), while the weft yarn consisted of 100% polyester obtained from Elephant Star (Indonesia). The fabric used in this study was a combination of polyester and rayon viscose fibers. To design and create this double-layered fabric, computer software called WiseTex and TexGen were used. The finished fabric had 70 horizontal threads and 98 vertical threads per inch.

(b) Fabrication of textile-based sensors

The fabrication of textile-based sensors employed the direct coating method on a double-faced woven substrate with an areal dimension of 18 x 28 mm and a substrate thickness of 0.82 mm. During the coating process, carbon paste was applied for the working and counter electrodes, while the reference electrode was coated with Ag/AgCl paste using the direct coating method assisted by molding. In the multi-layer textile-based concept, the quantity of carbon paste applied to the substrate was adjusted to 0.08 grams for one layer of the working electrode and 0.02 grams for the counter and reference electrodes. In the single-layer textile-based concept, the amounts of carbon and Ag/AgCl paste used were only approximately one-third of those used in the multi-layer textile-based concept. The curing process for the electrode paste was carried out at a temperature of 110°C for a duration of 20 minutes. Subsequently, the resulting sensor was coated with a hydrophobic barrier to limit the wetting area by the analyte.

(c) Sensor Performance

The performance measurement process of the sensor commences with the application of 0.15 mL of glucose oxidase enzyme solution (0.19 units) on the surface of the working electrode of the textile-based sensor. For the conventional commercial screen printed carbon electrode (SPCE) sensor, the amount of glucose oxidase enzyme solution used is 0.05 mL (0.06 units). Subsequently, a drying process is carried out at room temperature for a duration of 4 hours. The measurement process is conducted using the PalmSens4 device through the chronoamperometry method at a voltage of 0.55 V with a duration of 300 seconds and an interval time of 0.5 seconds.

In this study, the analyte used is a glucose solution with concentrations of 10 mM, 20 mM, 30 mM, 40 mM, and 50 mM, each repeated five times for every concentration variation. The measured current begins 30 seconds after achieving a relatively stable value, and then the average is calculated for all concentration variations. The calculated averages from all variations are then summarized, followed by regression analysis and linear equation calculations to determine the sensitivity and linearity of the sensor. As a comparison to the performance of the textile-based sensor, measurements are also conducted using the same method with a commercially available screen-printed carbon electrode sold in the market.

(d) Sensor Durability

The durability assessment of the sensor in this study encompasses its stability under repeated folding and resistance to bending. Measurements were conducted using the chronoamperometry method at a voltage of 0.55 V with a duration of 300 seconds and an interval time of 0.5 seconds in a glucose solution with a concentration of 30 mM, totaling 0.2 mL. The measured current, stabilized after 30 seconds, is then subjected to average calculation for all concentration variations. The calculated average current densities from all variations are summarized, followed by the computation of the coefficient of variation to ascertain the stability of the measurement results.

The durability of the sensor against repeated bending is assessed by subjecting it to repeated folds at a 90° angle, with variations in intensity at 0, 5, 10, 25, and 50 times before performing chronoamperometry measurements. The durability of the sensor against bending is evaluated by measuring the current density of a 0.2 mL glucose solution with a concentration of 30 mM while applying bends to the sensor. Different bend angles are applied in this process, namely: 0°, 45°, 90°, 135°, and 180°, with the assistance of two metal plates and a protractor.

3. Results and Discussion

(a) Fabrication of textile-based sensors

The fabrication results of textile-based sensors single-layer concepts through the direct coating method exhibit electrode patterns resembling those of the original molds. However, there is a notable alteration in the spacing between electrodes, particularly between the reference and counter electrodes, changing from an initial 2 mm distance to 4 mm. This modification is attributed to electrode paste spillage during the coating process, leading to potential short circuits and influencing the resultant sensor performance. One potential solution to minimize this issue is considering the use of printing methods capable of seamlessly integrating each element onto the fabric surface [11].

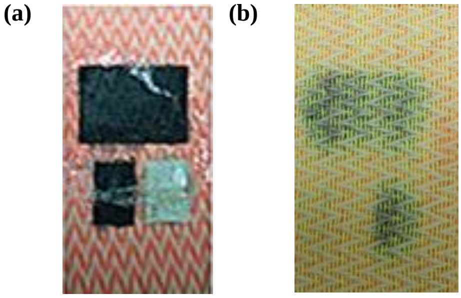

Figure 1 Single-layer textile based sensor; (a). Front side; (b). Back side

(b) Sensor Performance

The measurement process was conducted using glucose solutions with concentrations ranging from 10 to 50 mM at a voltage of 0.55 V. To compare the performance of the textile-based sensor, measurements were also performed using the same method with a commercially available Screen Printed Carbon Electrode (SPCE). Chronoamperograms of glucose solutions at various concentrations on SPCE and STBS can be observed in Figure 2 (a-b). Based on the chronoamperogram measurement results for SPCE in Figure 2, it is evident that the glucose solution with a concentration of 10 mM exhibits the lowest average current value, while the solution with a concentration of 50 mM shows the highest average current value. There is an increase in the average current value with the rise in the concentration of the glucose solution used. The higher the concentration of the glucose solution, the higher the average current value generated. This phenomenon is attributed to the increase in the number of electrons during the reaction.

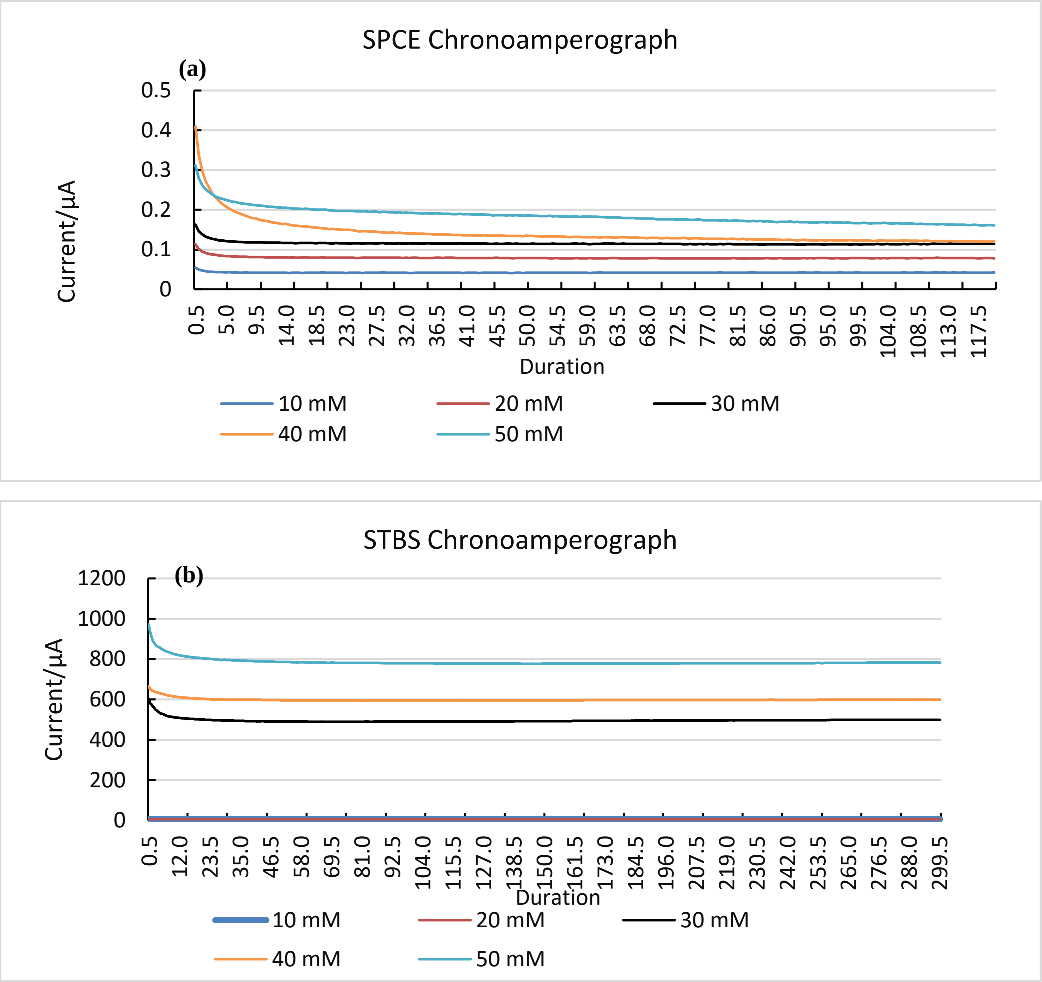

Figure 2 The chronoamperograms of glucose solutions at various concentrations: (a). Screen-printed carbon electrode and; (b). Single-layer textile based sensor.

The subsequent potentiostat instrument indicates the presence of current during chronoamperometry measurements of glucose solutions at concentrations ranging from 30 to 50 mM. The chronoamperometry measurement results for glucose solutions with concentrations of 30 - 50 mM exhibit a phenomenon consistent with that observed in measurements using commercially available conventional sensors (SPCE). The glucose solution with a concentration of 30 mM has the lowest average current value, while the solution with a concentration of 50 mM has the highest average current value. The increase in glucose concentration leads to an increase in the detected current during measurements [11].

(c) Folding Durability

Sensors utilizing a non-invasive method are expected to exhibit good durability against repeated folding, especially for applications involving continuous systems. This is because the sensor application process occurs simultaneously with user activities in parallel. These activities may cause the sensor to fold repeatedly to adapt to the type and duration of the performed activities. Therefore, a sensor with good flexibility is needed to maintain measurement performance throughout the application.

The chronoamperogram of glucose solutions after repeated folding in various variations can be observed in Figure 3 (a-b). Based on the chronoamperogram of the SPCE sensor in Figure 3 (a), there is instability after the SPCE sensor undergoes repeated folding 10 times. Subsequently, there is a decrease in the average current value after the SPCE sensor undergoes folding 25 and 50 times, where the average current value approaches zero. This phenomenon may be attributed to the decrease in the functionality of the electrode on the SPCE sensor caused by surface cracks in the folded area. These cracks result from the detachment of carbon paste and Ag/AgCl from the polypropylene solid substrate of the SPCE sensor.

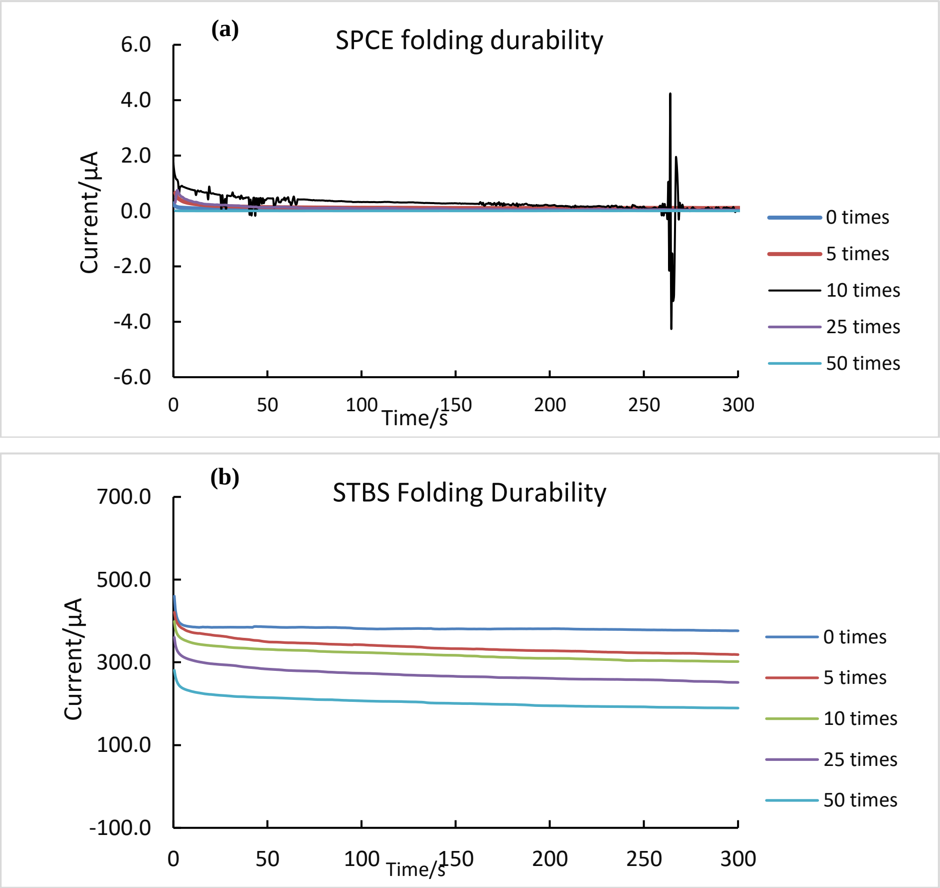

Figure 3 The chronoamperograms of glucose solutions at various folding repetition: (a). Screen-printed carbon electrode and; (b). Single-layer textile based sensor.

A different phenomenon occurs in the durability measurement of STBS against repeated folding. Based on the chronoamperogram in Figure 3 (b), the entire graph of the measurement results appears stable. The detected average current value in the STBS, which has not undergone repeated bending, is the highest, and this value gradually decreases with the occurrence of repeated bending in the SPCE. This could be attributed to the partial detachment of electrode paste and changes in fiber position during the folding process [13].

(d) Bending Durability

The chronoamperogram in Figure 4 (a-b) demonstrates the curve shapes are relatively similar between the SPCE and STBS sensors. The difference is noticeable in the SPCE sensor, where during the durability testing against bending, there is a sharp increase in current at the beginning of the test, persisting until the 88th second when applying a 180° bending angle. This phenomenon may be attributed to the stacking of the analyte at the initial bending of the SPCE sensor, and the proximity of the electrodes leading to a faster electron transfer process compared to the earlier stages.

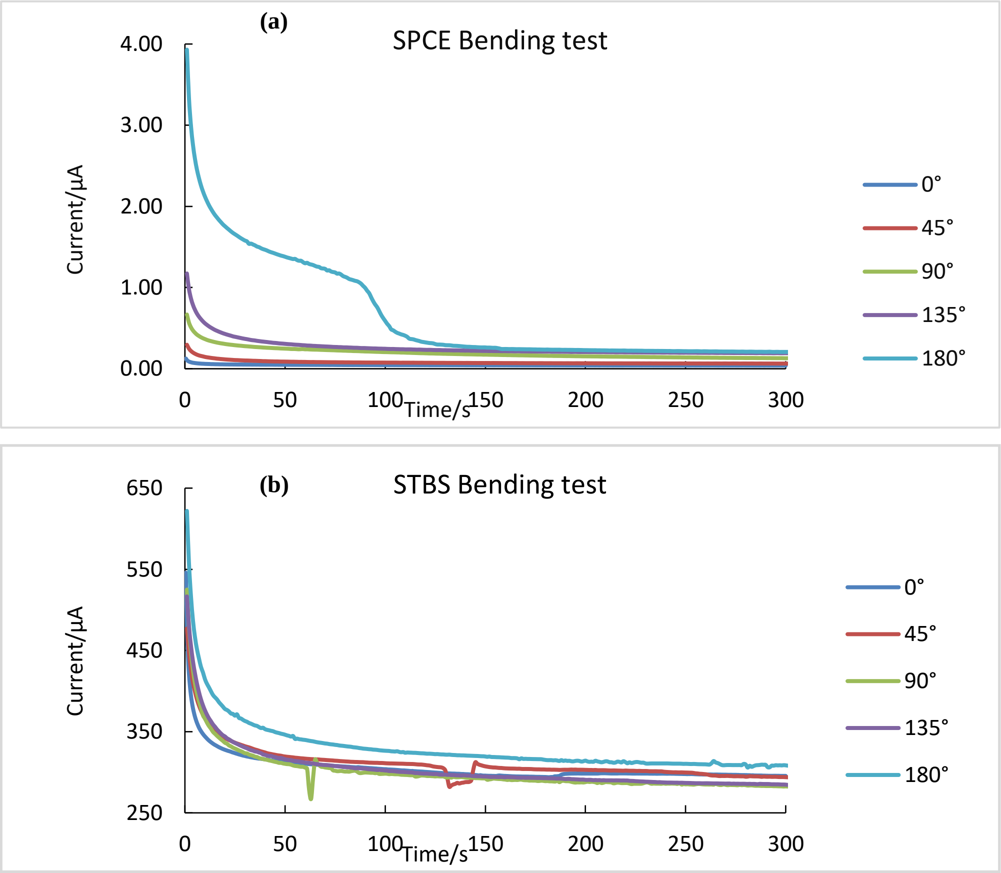

Figure 4. The chronoamperograms of glucose solutions at various bending angles: (a). Screen-Printed Carbon Electrode and; (b). Single-layer textile based sensor.

The calculation results indicate that the coefficient of variation for SPCE is 19 times higher than the coefficient of variation for STBS. The coefficient of variation for the durability of SPCE against bending reaches 67.28%, while the coefficient of variation for the durability of STBS against bending is 3.55%. Based on these results, it can be inferred that in this study, STBS exhibits better stability compared to SPCE. This is attributed to the fact that the current values recorded by the potentiostat instrument at various bending angles in STBS are relatively more homogeneous than the current values generated by the SPCE sensor.

4. Conclusion

The single-layer textile-based sensor examined in this study exhibits enhanced stability compared to traditional SPCE sensors. This improved stability is linked to the textile substrate, which provides increased flexibility and durability. Additionally, the single-layer textile-based sensor demonstrates greater sensitivity when compared to screen-printed carbon electrodes. The increased sensitivity observed is a notable advantage, suggesting that the multi-layer concept has the potential to improve the sensor's capacity to detect and react to changes in the target analyte. This positions it as a promising option for applications requiring both high sensitivity and stability.

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Conflict of interest: The authors declare that there is no conflict of interest.

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