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Chemically reduced graphene oxide-based dry electrodes as touch sensor for electrocardiograph measurement

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ABSTRACT

This article presents the fabrication of a flexible, conductive, and chemically reduced graphene oxide (CRGO)-based dry electrodes for electrocardiograph signal measurement using touch sensing mechanism. We designed the dry electrode sensor to eliminate the noise and inconvenience associated to the use of electrolytic gel. CRGO electrodes were used as touch sensors for obtaining high quality biopotential signal and human fingers were utilized to maintain good contact to enable minimizing the skin-electrode contact impedance. The surface resistivity of the CRGO electrode was found 28 Ω/square. The proposed method is a simple and cost-effective fabrication process for a flexible conducting CRGO-coated nylon membrane for electrocardiograph (ECG) signal monitoring application.

1. Introduction

Cardiovascular disease is significant amongst the most serious threats to human health, and many heart diseases usually occur suddenly and unexpectedly. This causes not only a great deal of anxiety for the patients, but also the chance of endangering their life. Several groups have created prototype systems for these purposes. Two types of electrodes are used with ECGs: dry electrode and wet electrode. The Ag/AgCl electrode is a wet electrode that is generally utilized in normal ECG measurement because of its low skin-electrode contact impedance, which assists accurate signal recording. However, it is generally not appropriate for long term observation because it can cause skin allergies resulting in skin and dermal irritation. Alternative electrodes are being investigated that could meet the requirements for long term ECG monitoring. These are known as dry electrodes that are effectively wearable [1], where accurate vital electrophysiological signal recording is desired. These electrodes are made of metal, conductive reduced graphene oxide (RGO) paper, conductive rubber or conductive fabric, and are body compatible and convenient for daily wear. They can maintain good contact with the skin even during movement, and are thus appropriate for use in ambulant circumstances such as home-care and exercise monitoring. Polydimethylsiloxane (PDMS)-based electrodes are also broadly utilized for different applications as part of an ECG acquisition system where adaptability and stretchability are required. Flexible PDMS dry electrodes [2] have been proposed for ECG monitoring application. Flexible ECG electrodes with textile integration [3–5] have been proposed in a number of studies. A dry conductive textile was prepared by dipping nylon fabric into a RGO solution, followed by a thermal treatment that allowed the formation of a conformal coating of conductive graphene layers around the fabric [6]. In this study, conductive CRGO paper was used to build the electrodes for ECG recording from human fingers. CRGO paper fits well to the contours of human fingers, enhancing the skin-electrode contact. One of the advantages of this electrode is that it does not require gel [7,8,14,15], which can cause skin aggravation. The performance of the fabricated electrode matches that of the conventional Ag/AgCl electrode.

1.1. Skin-electrode interface equivalent circuit

The equivalent circuit model of the skin/electrode interface for standard wet and dry electrodes is shown in Fig. 1(a) and (b). $Na+$, $K+$, and $Cl-$ ions produce a biopotential due to the combined divergence between the internal and external conditions of a cell. Biopotential electrodes [9,10] can pick up the biopotential from the skin by conducting a current across the interface between the skin and the measurement circuit. Biopotential electrodes work as a transducer that changes an ionic current into an electronic current. So, when a standard wet electrode is in operation, a half-cell that comprises a metal electrode part is formed. The coupling layer between the skin and the electrode can be described as a resistor and a capacitor in parallel, respectively. In Fig. 1(a), R_d and C_d represent the impedance and the capacitance of the electrode, respectively. R_e denotes the RC element of the epidermis. C_e and R_u denote the RC elements of the dermal layers. The potential difference E_{se} is due to the difference of the ionic concentration across a semi-permeable membrane, which is given by the Nernst equation [4]. E_{he} denotes the electrode potential and R_s represents the effective resistance of the conductive gel. In contrast with the conventional wet

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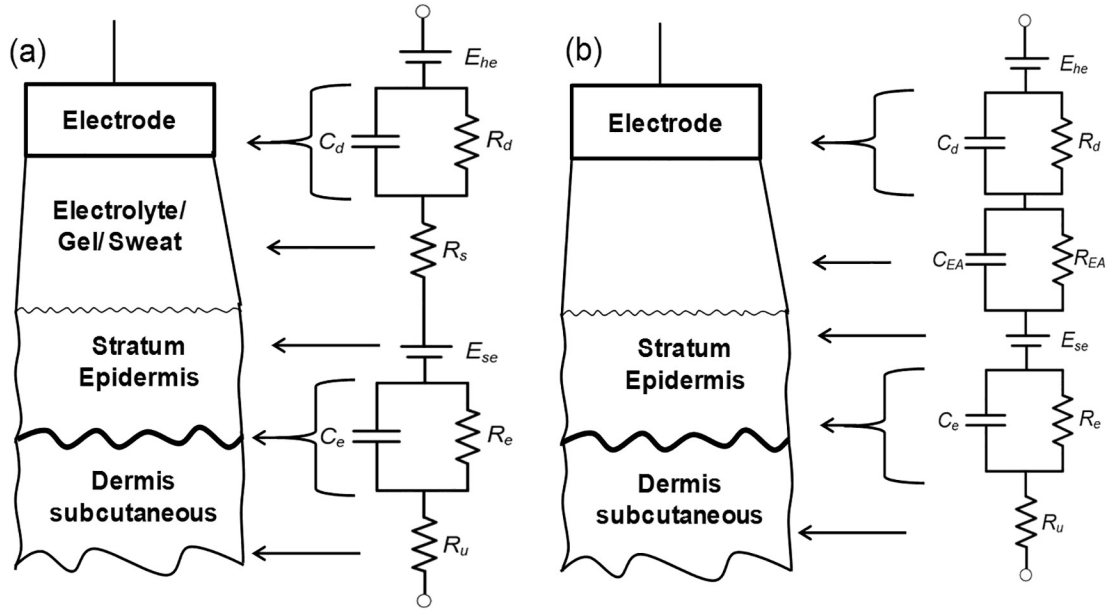

Fig. 1. Equivalent circuit: (a) wet electrode and (b) dry electrode.

electrodes, the dry electrode is capable of the following behaviors: (1) a functionality without gel, and (2) the provision of a capacitive behavior at the interface. The equivalent circuit model for the dry electrode/skin interface consists of C_{EA} and R_{EA}, which are connected in parallel, as shown in Fig. 1(b).

2. Materials and methods

2.1. Fabrication of electrodes

The modified Hummers method [6] was used for preparing the graphite oxide. Briefly, 6 g graphite powder, 5 g of $K_2S_2O_8$, and 5 g of P_2O_5 were added in 25 ml of 98% H_2SO_4 solution. The solution was mixed continuously at 80 °C for 4.5 h. It was then cooled to room temperature and diluted with 1 l of de-ionized (DI) water and left overnight. The pre-oxidized graphite was filtered using the DI water and dried in a vacuum oven at 50 °C. Subsequently, 3 g of pre-oxidized graphite was added to 120 ml of 98% H_2SO_4 solution in a flask immersed in an ice bath. Simultaneously, 15 g $KMnO_4$ was added gradually while stirring to prevent rapid aggregation of the evolved heat. The mixture was then stirred at 40 °C for 0.5 h and 90 °C for 1.5 h with the drop wise addition of 250 ml of water, which was incubated at 105 °C for 25 min and stirred at room temperature for 2 h. To terminate the reaction, 0.7 l of water and 20 ml of 30% (w/w) H_2O_2 were added. Subsequently, the product was filtered and washed with 3 M HCl solution, and repeatedly washed with water until achieving a neutral pH value. Before drying the filtered product at 60 °C in a vacuum oven, chemically modified graphite oxide (CGO) platelets were collected. 100 mg of CGO were dispersed in 60 ml of ethylene glycol, followed by sonication for 1.5 h. 80 ml of water was added to the solution, followed by stirring for 1 h. 540 mg of sodium borohydride ($NaBH_4$) was slowly added and the mixture was then heated at 110 °C for 2 h while stirring. The reaction mixture was filtered, washed with doubly distilled water, and then dried under a vacuum oven at 100 °C after the reduction reaction had finished. We used 0.2 μm nylon membrane filter paper with a vacuum pump to accelerate the filtration after chemical conversion of GO to RGO. After filtration, the nylon membrane filter paper was annealed using a hot plate at 90 °C for 15 min. Fig. 2 shows the process of making a dry CRGO electrode. The prepared CRGO paper was connected to the ECG cable using steel and a nickel-coated connector with the ECG signal acquisition system. Fig. 3(a) shows the scanning electron microscope (SEM) image of blank nylon membrane filter paper. In Fig. 3(b), well-defined sheets that are uniformly deposited over the surface of the nylon membrane paper can be seen. CRGO paper is therefore obtained, exhibiting surface resistivity of as low as 28 Ω/square, as shown in Fig. 3(c). The surface resistivity was calculated using a four-point probe (RC2175, EDTM).

This demonstrates an increase in the conductivity, significantly outperforming the chemical treatments. To examine the adhesion between CRGO and nylon membrane, a simple adhesion test was done using scotch tape as shown in Fig. 3(d). It is clearly shown that the scotch tape could be easily released manually from the CRGO electrode without damaging the material. High resolution X-ray photoelectron spectroscopy (XPS) scans from the carbon region (C1s) were performed on the paper materials studied. The most prominent components were deconvoluted with the help of a fitting program and the obtained spectra are shown in Fig. 3(e). Deconvolution of C1s in the CRGO paper (Fig. 3e) revealed the presence of a strong peak at ~284.61 eV, corresponding

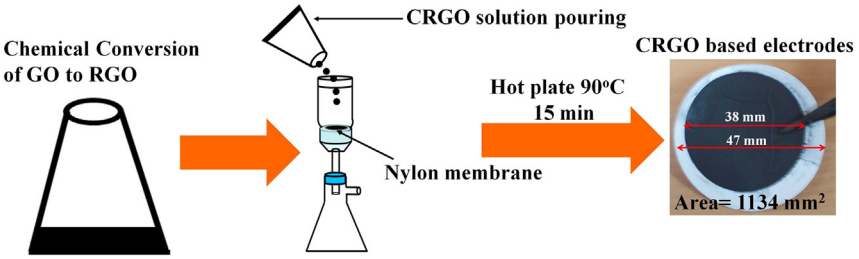

Fig. 2. Fabrication process of the CRGO dry electrodes.