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Flexible microneedle array electrode using magnetorheological drawing lithography for bio-signal monitoring

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ABSTRACT

Monitoring and timely intervention are extremely important in the continuous home care. Microneedle array electrode (MAE) have been employed for the long-term bio-signal monitoring without skin preparation. We developed a novel magneto-rheological drawing lithography (MRDL) method to cost-effectively fabricate a flexible micro-needle array electrode (FMAE) for the wearable bio-signal monitoring. Flexible substrate may match closely with curved skin and maintain a stable interface between skin and electrode. The formation mechanism of microneedle array (MA) by MRDL and bio-signal recording performance of FMAE were investigated. MA can be one-step drawn from the droplet array of curable magnetorheological fluid under the assist of external magnetic field. Ti/Au film was coated on the surface of solidified MA to insure the conductivity and compatibility of FMAE. 36-FMAE consists of 6 × 6 microneedles with an average height of 600 μm and an average tip radius of 12 μm. FMAE with 36 needles (36-FMAE) shows a better bio-signal monitoring performance in some specific situations compared with flexible dry electrode (FDE) and commercial Ag/AgCl electrode. Electrode-skin interface impedance (EII) measured by 36-FMAE is the lowest at a given low input frequency and the amplitude of electrocardiography (ECG) and electroencephalography (EEG) signals recorded by 36-FMAE is the largest. 36-FMAE can collect more distinguishable features and weaken the effect of motion artifact during the dynamical ECG recording. Therefore, 36-FMAE is a promising sensor for the wearable bio-signal monitoring in home care.

1. Introduction

Wearable and flexible sensors have been gaining large interest over the last decade in the modern home monitoring and diagnose [1,2]. Mobile and long-term monitoring of bio-signals, including electrocardiography (ECG), electromyography (EMG) and electroencephalography (EEG) signals, can improve the early diagnosis of diseases and ongoing treatment for patients at home. For cardiovascular patients, it is important to monitor the heart attacks in real time by the continuous recording of ECG signals. For muscular dystrophy patients, it is helpful to estimate the curative effect by detection of EMG signals. For epilepsy patients, it is useful to identify epilepsy events by the long-term recording of EEG signals. Therefore, it is necessary to provide a long-term wearable electrode for the continuous monitoring of bio-signals.

Currently, typical biomedical electrodes, including wet electrode and dry electrode, have been widely employed to capture bio-signals. Wet electrode, such as gel based Ag/AgCl electrode, is the most common method. Conventional wet electrode requires skin preparation and gel usage to obtain low interface impedance, which limits its further applications in the long-term monitoring of bio-signals [3–5]. Dry electrode is another alternative method without gel usage, which shows promising feature for the continuous recording. However, dry electrode has high interface impedance of stratum corneum layer and is sensitive to human motion [6,7]. To address above problem, microneedle array electrode (MAE), as an improved dry electrode, was introduced. MAE can easily pierce through the stratum corneum layer and directly capture bio-signals in the living epidermal layer, eliminating high impedance of stratum corneum [8]. The skin-electrode interface is more stable due to skin penetration, weakening the effect of motion artifact [9–11]. However, most of MAE are usually based on rigid substrates that are not conformal and instable contact to curved and moved human skin [5,12]. Meanwhile, flexible microneedle array electrode (FMAE) could mechanically couple with the curved skin well,

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provide robust contact, and further minimize the motion induced artifacts [3,5,13]. Therefore, FMAE is a promising flexible sensor for the long-term bio-signal monitoring.

Microneedle array (MA) is the key component of MAE and its typical fabrication approaches includes lithography with etching [14–18], micromolding [4,12,19], laser machining [20,21], 3D printing [22], thermal drawing [11] and magnetization-induced self-assembling [10,23] and so on. Most MA are fabricated on rigid substrate, such as silicon, stainless steel, copper, polymethyl methacrylate (PMMA), poly (lactic-co-glycolic acid) (PLGA) and so on. Reports on fabrication of MA on the flexible substrate are very less due to its complex fabrication process. Kim [12] proposed a curved MAE for robust long-term measurements, high selectivity, and easy applicability. The MA was fabricated by a micromolding process, but its curved substrate designed to fit the curvature of skin was rigid. Wang et al. [5,13] developed a flexible parylene-based MAE for the long-term monitoring of bio-potentials. However, its fabrication process, including thermally oxidize, reactive-ion etching, deposition of parylene films, lift-off technique, sputtering and so on, was extremely complex and required expensive equipment. Srivastava et al. [3] presented a flexible polymer photoresist (SU-8) based MAE for sensing of bio-signals. The MA was fabricated by the UV maskless lithography, which is suitable for large scale production. But it requires expensive and sophisticated equipment located in clean rooms. Therefore, development of a cost-effective fabrication process of FMAE for bio-signal monitoring is still a challenge.

We present a novel approach named magneto-rheological drawing lithography (MRDL) to efficiently fabricate MA on the flexible substrate molding-free. This approach is an additive technique directly and rapidly drawing 3D MA from the droplets of curable magnetorheological fluid (CMRF) under the assist of external magnetic field. It is extremely simple and suitable for large scale facile production. The solidified MA coated with Ti/Au would be assembled as a polyimide based FMAE. The recording performance of bio-signals, including electrode-skin interface impedance (EII), ECG, EMG and EEG, would be measured in comparison with commercial Ag/AgCl wet electrode and flexible dry electrode (FDE).

2. Materials and methods

2.1. Design and fabrication of FMAE

(1) CMRF preparation: epoxy novolac resin (Weiyi Metallography Experiment Instrument Co., LTD, China) and iron particles with an average diameter of 1 μm (Naiou Nano technology Co., Ltd, China) were purchased. CMRF was prepared as: firstly, epoxy novolac resin was uniformly mixed with iron particles with a mass ratio of 1: 0.5. Subsequently, the mixture was pre-polymerized at 80 °C for 3 min.
(2) FMAE design: FMAE was designed and its dimensions are presented, as shown in Fig. 1(a). The pattern of flexible PCB was designed by Altium Designer (Altium Information Technology Co., LTD, China) and fabricated with the polyimide substrate. The rectangular pads can be connected with a multiway switch. By adjusting of the multiway switch, one microneedle electrode (1-FMAE), nine microneedles electrode (9-FMAE), eighteen microneedles electrode (18-FMAE) and thirty-six microneedles electrode (36-FMAE) can be turned to record the bio-signals.
(3) FMAE fabrication: A MRDL setup was self-developed for the rapid fabrication of MA, as shown in Fig. 1(b). The whole fabrication process of FMAE is presented in Fig. 1(c). Firstly, the pillar tips were firstly coated with the droplets as the pillar tips were dipped in a pool of curable magnetorheological fluid (CMRF). The diameter of copper pillar is 0.7 mm. Secondly, the pillars were moved toward the substrate at a speed of 1.5 mm/s by a linear motor (C-884, PI, German). The droplets were compressed on the substrate for 1 s. Subsequently, pillar array was drawn back at a speed of 1.5 mm/s and stopped at a distant of 12 mm away from the substrate. A liquid MA was formed on the substrate under an external magnetic field intensity of 100 mT. The fabrication process was carried out at room temperature. The liquid MA was pre-baked by hot air blowing at a temperature of 95 °C for 5 min. The fabrication process was monitored by an optical microscope. The pre-baked microneedle was further solidified in a vacuum oven at a temperature of 100 °C for 1 h. 20 nm Ti film and 200 nm Au film were uniformly coated on the surface of solidified MA by the magnetron sputtering machine (MSP-3300, Jinshengweina Technology Co., Ltd, China) in sequence [13,22]. A FMAE was fabricated and ready for bio-signal recording. A FDE without any MA on the substrate was also fabricated by following above process.

2.2. Bio-signal monitoring

In order to better evaluation of the bio-signal monitoring performance of FMAE, EII, ECG, EMG and EEG signals were recorded in comparison with conventional wet electrode (Ag/AgCl electrode, JK-1, Junkang Medical Supplies LTD., CO, China) and FDE. The medical adhesive tapes with a size of 30 mm × 15 mm were used to keep the FMAE and FDE close on the skin. We tried our best to measure the bio-signals under the same conditions. The signals from these different electrodes were recorded in sequence. Three healthy volunteers from 23 to 27 years old were tested and repeated at least for five times. The detailed test procedures of EII, ECG, EMG and EEG was well descripted in our previous work [10,11]. This study was approved by the ethics committee of the Work Injury Rehabilitation Center of Guangdong Province (Approval No. AF/SC-07/2016.29). All volunteers provided written informed consent.

2.2.1. EII test

A two-electrode measurement method was used to record EII using a precision impedance analyzer (Agilent E4980A LCR Meter, Palo Alto, CA, USA). Two electrodes were placed on the left inner forearm with a distance of 5 cm.

2.2.2. ECG test

The ECG100C module of Multipurpose Polygraph (MP150, BIOPAC, Goleta, CA, USA) was employed to record static and dynamic state ECG signals using the standard I-lead method. Two working electrodes were placed on the right wrist and left wrist, respectively. The ground electrode was on the right ankle. The subjects were asked to lie in bed during the static ECG test and walk on a treadmill at a uniform speed of 3 km/h during the dynamic ECG test.

2.2.3. EMG test

The EMG signals of biceps brachii were recorded by the differential method. Two working electrodes were placed on the biceps brachii with a distance of 2 cm, and the ground electrode was placed on the elbow.

2.2.4. EEG test

An EEG100C module of Multipurpose Polygraph (MP150, BIOPAC, Goleta, CA, USA) was employed to record EEG signals using the unipolar connection method. One working electrode was placed on the standard position (Fp1) of the 10–20 system. Another working electrode and the ground electrode were placed on the left earlobe.