Available online at www.sciencedirect.com
ScienceDirect
Sensors and Actuators A 143 (2008) 423–429
SENSORS AND ACTUATORS A PHYSICAL
www.elsevier.com/locate/sna

Flexible polymeric dry electrodes for the long-term monitoring of ECG

Ju-Yeoul Baek [a,c], Jin-Hee An [a], Jong-Min Choi [b], Kwang-Suk Park [b], Sang-Hoon Lee [a,*]

[a] Department of Biomedical Engineering, Korea University, 126-1 Anam-dong 5ga, Sungbuk-gu, Seoul, 136-705, South Korea
[b] Department of Biomedical Engineering, College of Medicine, Seoul National University, 28 Yeongeon-dong, Jongno-gu, Seoul, 110-799, South Korea
[c] Department of Biomedical Engineering, Yonsei University, 234 Maeji, Heungeop, Wonju, 220-710, South Korea

Received 19 August 2007; received in revised form 15 November 2007; accepted 15 November 2007
Available online 24 November 2007

Abstract

In this paper, we present a novel polymeric dry electrode that (1) changes its shape in a way that supports the electrode's contact with the skin and (2) that does not cause skin irritations or allergic reactions. For a polymeric substrate of electrodes, we have used the elastomer poly(dimethylsiloxane), which is known to be inexpensive, biocompatible, and amenable to micro-molding, and to have excellent gas and water permeability. We have established a process by which one can deposit a metal layer on the PDMS substrate, etch the electrode patterns chemically and with good resolution, and package the electrode so that it is easily wearable on the forearm. We measured the impedance according to the frequency change and compared the results with those of Ag/AgCl electrodes. Afterward, we measured the ECG signal and investigated possible artifacts caused by motion. For the feasibility of long-term monitoring, we examined the influence of surface electrodes on the skin after 7 days of ECG monitoring. In conclusion, our PDMS-based dry electrode measured the ECG signals with comparatively good fidelity, but showed better skin compatibility after long-term tests. We expect that our method for the production of PDMS-based dry electrodes will be broadly applicable to the field of ubiquitous biosignal monitoring.

Keywords: Dry electrodes; PDMS; Long-term monitoring; ECG

1. Introduction

The rapid development of telecommunications during the last few decades has made it possible to monitor personal health in the absence of individual care and regardless of patients' locations and status (e.g.: sleep or exercise) [1–5]. The development of special bioelectrodes is one of the most significant factors in the realization of such ubiquitous health care. These bioelectrodes help people in clinical field measure biosignals, such as with an electroencephalogram (EEG), electromyogram (EMG), and electrocardiogram (ECG), with high fidelity and to conduct long-term (1 week) monitoring without damage to the skin. The most commonly used bioelectrodes in the clinical fields are of the gel type silver/silver chloride (Ag/AgCl) type [6], and these electrodes are simple, lightweight, reliable, and cost effective. However, these electrodes have the limitation that the long-term use of them may irritate the skin.

Because of this limitation, researchers have tried to discover alternatives to Ag/AgCl electrodes that would still meet the requirements of the long-term monitoring of biosignals. One of the alternative methods is the use of dry electrodes. Even though positive results of the dry electrode have been published [7,8], known dry electrodes, most of which were made on hard or stiff substrates, often cause the following problems: (1) the contact impedance to the skin is higher than that of gel electrodes, (2) the contact area varies owing to its rigidness during the motion, and (3) the electrode causes damage to the skin over the long-term. To overcome these limitations of dry electrodes, researchers have conducted many trials such as invasive mechanical abrasion of the skin, carbon-nanotube-based electrodes [9], and foam electrodes [10]. These methods have contributed partially to the quality improvement of dry electrodes but have their own respective shortcomings. In this paper, we present a novel polymeric dry electrode that (1) changes its shape in a way that supports the electrode's contact with the skin and (2) that does not cause skin irritations or allergic reactions.

For a polymeric electrode substrate, we have used the elastomer poly(dimethylsiloxane) (PDMS), which is known to be inexpensive, biocompatible, and amenable to micro-molding,

* Corresponding author. Tel.: +82 2 920 6457; fax: +82 2 921 6818.
E-mail address: dbiomed@korea.ac.kr (S.-H. Lee).

0924-4247/$ – see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.sna.2007.11.019

and to have excellent gas and water permeability [11,12]. In spite of the many advantages of the PDMS substrate, a big challenge has been the stable depositing of metal layers on the surface of the PDMS. Even more difficult has been the effort to pattern the deposited metal layer via a chemical etching process.

Here, we have established a process by which one can fabricate a dry electrode on a PDMS substrate and can package the electrode so that it is easily wearable on the arm. Using diverse tests, we evaluated the functions of a dry electrode. We measured contact impedance according to frequency changes and compared the results with those of wet electrodes. Afterward, we measured the ECG signal and quantitatively investigated the motion artifact through the treadmill test. To determine the feasibility of long-term monitoring, we examined the influence of surface electrodes on the skin after 7 days of continuous ECG monitoring.

2. Materials and methods

2.1. Microfabrication of metal layer and base structure

The shape of the dry electrode is illustrated in Fig. 1a, and the substrate material is PDMS. For better contact of the metal pattern with the skin, the center has a convex shape protruding 1.5 mm upward, and the titanium (used as an adhesion layer) and the gold pattern is located on this center. We connected the Velcro to the end of the electrodes so that the electrode would be easily wearable around the forearm. Usually, it is difficult to build metal patterns on the curved PDMS surface. Therefore, we fabricated the electrode via two steps. First, we prepared an electrode layer and a base structure separately. Second, we bonded them to each other by following the following process, as in Fig. 1b: (1) we conducted an oxygen plasma treatment on the surface of both the electrode layer (backside) and the base structure, (2) we uniformly spread the PDMS precursor on the treated electrode layer, and (3) we bonded the electrode layer to the base structure and thermally cured it at 80 °C for 2 h on the hot plate.

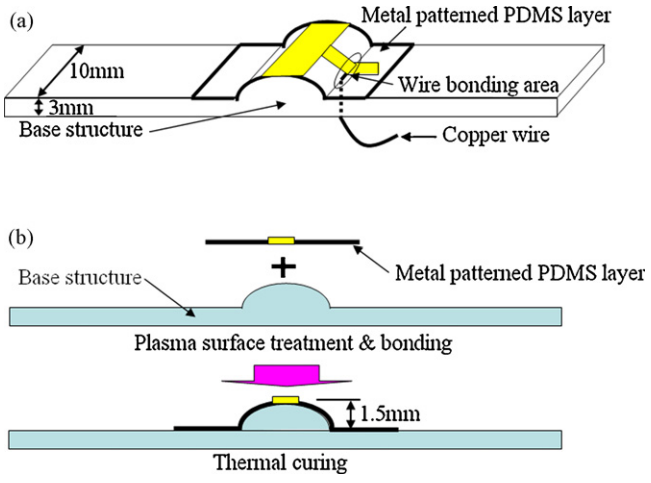

Fig. 1. (a) Schematic diagram of dry electrode, (b) bonding process of metal layer and base structure.

Table 1
Deposition conditions of metal layers (Ti and Au) on the PDMS surface

Parameter	Condition
Vacuum	Less then 1.5×10^{-6} torr
Deposition rate of Ti	0.5–3 Å/s
Thickness of Ti	50–300 Å
Deposition rate of Au	0.5–3 Å/s
Thickness of Au	500–3000 Å
E-beam focusing size	3 mm Φ
E-beam power	
Voltage	7.5 kV
Current	0–100 mA
Source to substrate distance	40 cm
Substrate Temperature	Non-heated

We fabricated the electrode layer according to the following process. First, we fabricated the thin PDMS substrate by the spin-coating method. Onto the 3 in. silicon wafer, we poured the 10:1 mixture of the PDMS pre-polymer and the curing agent (Sylgard 184 silicone elastomer kit, Dow Corning, Midland, MI). Then, we spin-coated (speed: 150 rpm) the mixture for 30 s and baked it for 2 h in the vacuum chamber at 120 °C. For 30 s, we exposed the surface of the PDMS layer to the oxygen plasma ion. Onto this PDMS surface, we e-beam evaporated the Ti (300 Å) and Au (3000 Å), respectively; the deposition conditions are summarized in Table 1. We then created the electrode metal patterns according to the following process: (1) we spin-coated the positive photoresistor (PR, AZ 5214, Clariant) on the e-beam-evaporated metal layer at a speed of 5000 rpm and baked it for 1 min at 110 °C, (2) we radiated the masked UV (365 nm, 300 mW) light on the PR-coated metal layer, (3) we developed the UV-exposed PR using the PR remover (AZ 300MIF, Clariant), (4) we applied an Au etchant (aqua regia) and a Ti etchant ($HF:HNO_3:H_2O = 1:2:7$ vol.%) to remove the metal layer unmasked by the PR, and (4) we removed the remnant PR on the metal pattern by using the PR stripper (AZ 400T, Clariant). We fabricated the base structure by using a UV-LiGA mold inserter. Onto the mold insert, we poured the 10:1 mixture of the PDMS pre-polymer and the curing agent and cured it for 2 h at 80 °C.

2.2. Packaging and wire connection

We fabricated the forearm-wearable electrode by connecting the Velcro to the PDMS electrode. We bonded the Velcro to the end of PDMS electrode by using the PDMS precursor as glue. The wire connection in the PDMS electrode is very challenging because the conventional methods such as wire bonder or soldering are almost impossible. We connected the wire to the metal pattern by using conductive glue, and we encapsulated the contact region (bonded region with conductive glue) with the PDMS precursor. The details of the wire bonding process are as follows: (1) we punctured a small hole and introduced a wire through this hole, (2) we bonded the end of this wire onto the gold pattern using conductive glue, and (3) we encapsulated the bonded region

using the PDMS precursor to maintain the stable connection.

2.3. Evaluation of the electrode via impedance and ECG measurement

We measured both the impedances of our electrode and solid gel type electrode (3 M red dot foam monitoring (Ag/AgCl) electrode 2237, 3 M Health Care) by using an impedance analyzer (Analytical 1260A, Solartron). Both the Ag/AgCl electrode and our polymeric electrode were placed adjacent to each other on a person's forearm (their position was at the center of the lower arm's anterior surface (between the wrist and the elbow)), and both electrodes were attached so that the same distance (14 mm) separated their centers. Because of these electrode locations, the conditions for the two electrodes were equivalent and a fair comparison was possible. Each measurement was carried out soon after applying the electrode (within 30 s) and after removal of moisture. We recorded the impedances between each pair of electrodes according to the change of frequency (1 Hz–1 KHz). We measured the ECG signals using an ECG amplifier (ECG100C, BIOPAC Systems) and set the frequency range of the ECG filter between 0.5 and 100 Hz. Using a treadmill exercise, we investigated the effect that motion would have on the measurement of ECG. We attached our PDMS electrodes onto both of the subject's arms (at the center of each forearm). To minimize the artifact induced by the extension cable of the ECG amplifier, we fixed the cable onto the body. For long-term monitoring of ECG signals, the essential requirement is that there must be a minimization of electrode-derived skin damage. To examine the effect of electrodes on the skin, we continuously attached our polymeric electrodes onto the three candidates' forearms for 7 days. After 7 days of use, we investigated the influence of the surface electrodes on the skin that they covered.

3. Results and discussion

3.1. Fabrication and packaging

We successfully fabricated dry electrodes that had a PDMS-based surface, and Fig. 2 shows a picture of a constructed electrode wearable on the forearm. The Au (3000 Å) and Ti (300 Å) layers were deposited stably on the surface of the PDMS layer and the metal patterns were created effectively through the chemical etching process which has been previously described [13]. The minimum width of the metal patterns that can be created via our fabrication process was 10 μm, and this size is sufficiently small for the application of surface electrodes. We carried out the peel-off test using adhesive tape (3 M), and the patterns stably adhered to the PDMS surface. We did not experience the peeling of metal patterns during the short-term (a few hours) or the long-term (7 days) experiments. We successfully produced the base structure using a UV-LiGA mold inserter via the simple fabrication process. The electrode layer was uniformly bonded along the base structure's curvature. We bonded the wire firmly onto the metal pattern and observed no disconnection even in the long-term experiments. The inset of Fig. 2 shows a picture of an electrode wound around the forearm.

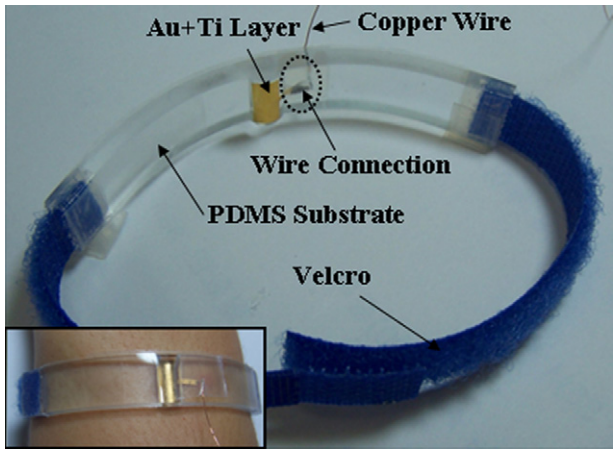

Fig. 2. Image of fabricated electrode (inset: picture of electrode wound around the forearm).

3.2. Measurement of impedances

Skin conductivity varies according to variations of the properties of either the stratum corneum or sweat glands and sweat ducts. In particular, the variation of impedance increased during dry-electrode use in response to changes in skin moisture, time, and pressure. To create reproducible measurements, we measured the impedance under the following conditions: (1) electrodes were positioned at constant locations at constant pressures, (2) measurements were carried out in 1 day, and (3) each measurement was carried out after removal of sweat. Under these conditions, we measured the impedances of both our electrode and the Ag/AgCl electrodes according to frequency changes, and Fig. 3 plots the results.

We conducted impedance measurements five times in each subject, and the results are shown in Fig. 3. The impedances of PDMS and Ag/AgCl electrodes (Fig. 3) are the mean value with standard deviation. We measured the impedance in relation to frequency ranges of 1 Hz–1 KHz. According to these measurements, the impedance of our PDMS electrode was much higher than that of the Ag/AgCl electrode at a

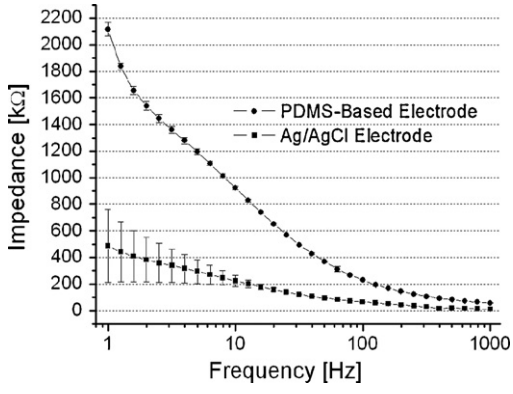

Fig. 3. Impedances according to the frequency changes (1 Hz–1 KHz).

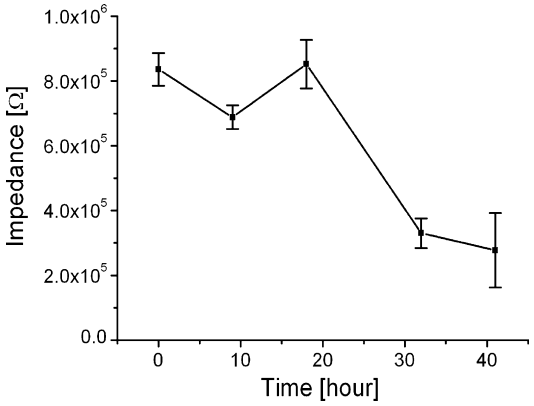

Fig. 4. 40 h of impedance monitoring (impedance values are the mean of five measurements).

frequency lower than 100 Hz. However, these impedance differences become almost similar at a frequency higher than 100 Hz. The trend of impedance according to the frequency change is similar to that of Karilainen's result [10]. We traced the impedance changes over the course of 40 h to monitor the long-term performance of PDMS electrodes. The result is plotted in Fig. 4. As expected, the impedance changed according to time because the environment and skin status, among other factors, are varied. However, the ECG signals were stably measured in spite of such impedance variations. In particular, the impedance declined significantly after the 24 h period, and we suspect that the skin under the electrode was slightly more moist.

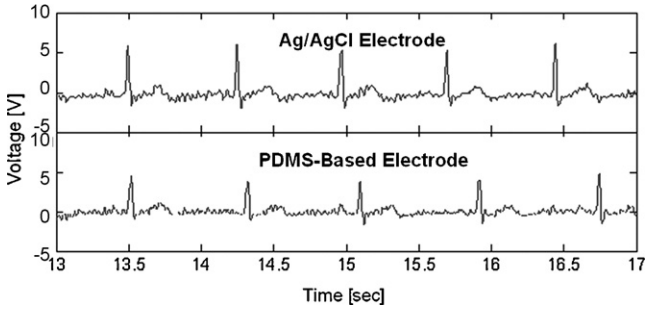

Fig. 5. ECG signals from the PDMS-based electrode and from the Ag/AgCl electrode.

3.3. Measurement of ECG signals

We measured the ECG signals using PDMS electrodes and Ag/AgCl electrodes at the lead I position, and the results are illustrated in Fig. 5. In comparison with the ECG signal of the PDMS and the Ag/AgCl electrodes, we discovered no significant differences between the two signals. The P, QRS, and T waves appeared clearly. The waveform of the ECG signal from the PDMS electrode was almost similar to that from the Ag/AgCl electrode.

We investigated the motion effects through the treadmill test and set the treadmill speed at 5 km/h. We measured the signals under both the rest status (which refers to the status existing just before the start of the exercise) and the exercise status. Fig. 6a and c illustrate the ECG signals of the PDMS electrode and of the Ag/AgCl electrode under the rest status just before exercise, and both signals were stable and almost the

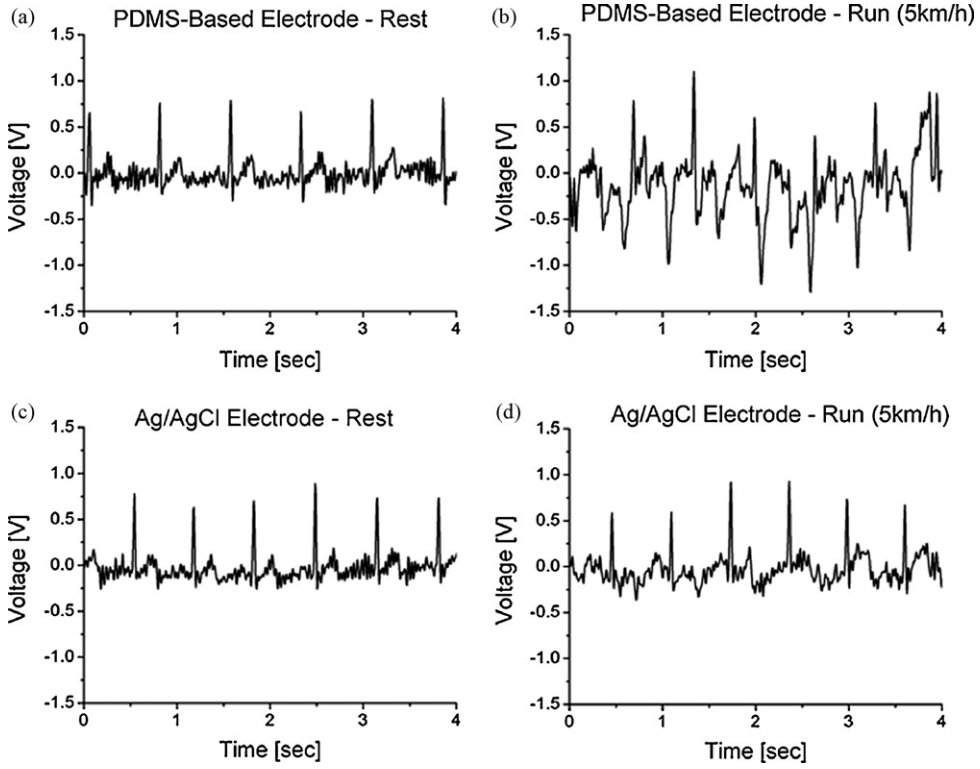

Fig. 6. ECG signals from the PDMS-based electrode and from the Ag/AgCl electrode before and after exercise (1 min after start of exercise, (a)–(d)).

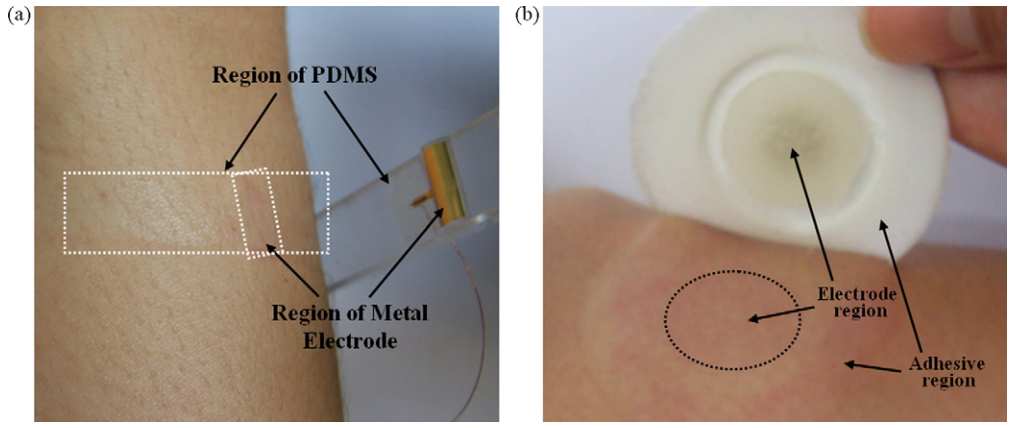

Fig. 7. (a) Skin under the PDMS-based dry electrode after 7 days of use, (b) skin under the Ag/AgCl electrode after 2 days of use.

same. We measured the signals of the PDMS electrode and of the Ag/AgCl electrode under exercise conditions, and Fig. 6b and d present the measurements. The motion had a greater effect on the ECG signal of the PDMS electrode than on that of the Ag/AgCl electrode; however, the R-wave was sufficiently detectable. The reason for this larger motion artifact is, in part, due to the high impedance of our dry electrode. Other reasons may be the unstable connection between the thin electrode wire and the thick ECG extension cable. This connecting area was shaken during the exercise period, even though we had fixed the cable and wire firmly to the body. We expect that the development of proper connection methods can reduce such motion artifacts.

3.4. Observation of the electrode's wearing effect

We examined the effect of surface electrodes on the skin. Three candidates attached both the dry electrodes and the Ag/AgCl electrodes to their forearm for 2 days. We investigated the influence of the two electrodes on the skin. In the case of the dry electrodes, we discovered no significant changes of the skin, such as irritation or itches, even under sweating conditions. However, the skin under the Ag/AgCl electrode turned red and became itchy in two subjects (Fig. 7b), so we stopped using the Ag/AgCl electrode in the experiment. We continued the experiment using our dry electrode for 1 week. Fig. 7a illustrates the status of the skin under our dry electrode, and we found no red

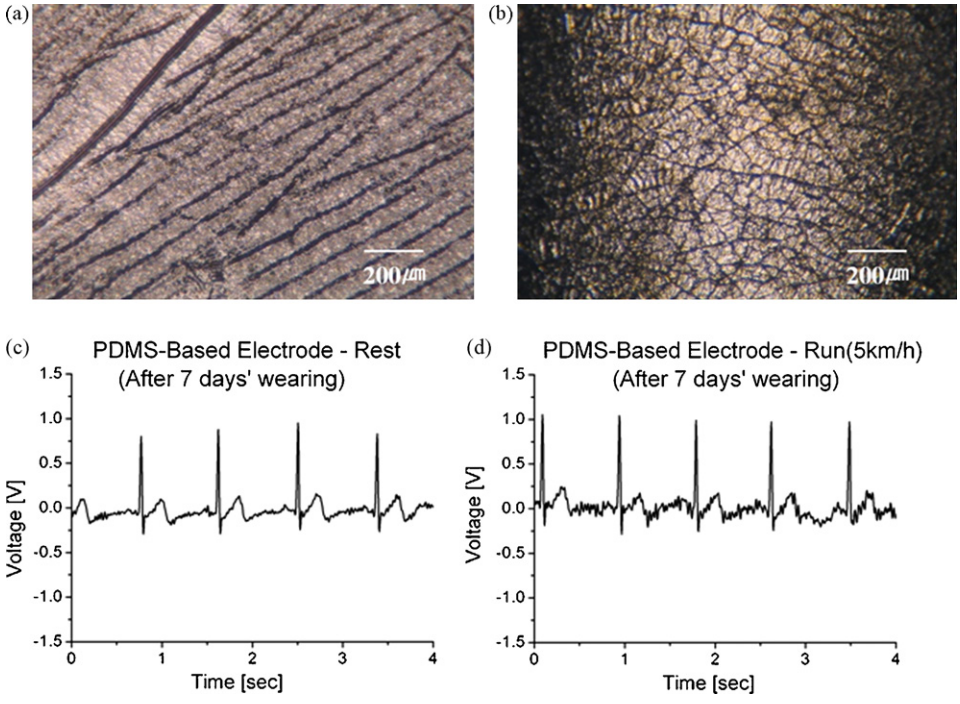

Fig. 8. Micrographs of electrode surface. (a) Before and (b) after 7 days of use. And ECG signals from the PDMS-based electrode after 7 days of use (c) and (d).

spots and subjects did not complain of itchiness. With our PDMS electrode, the subjects could shower or swim by disconnecting only the signal lines. To investigate the influence of 7 days of use, three subjects wore the electrode for 1 week continuously, after which time, the following two tests were carried out: (1) we examined the surface of the electrode with optical microscopy to observe the microcracks; (2) we measured the resistance of the electrode to examine if such microcracks influence the electrical properties of the electrode. Fig. 8a and b illustrates the micrographs of the electrode's surface before and after 7 days of use, and more minor cracks were observed in the worn electrode as expected. The electrical resistances of the two electrodes were measured, and they were 0.035 ± 0.021 and $0.252 \pm 0.126\,\Omega$, respectively. Even though a small increase of resistance was observed, the electrode still had good electrical properties after 7 days of use. The ECG signals were measured after the electrodes were worn for 7 days, and the result is plotted in Fig. 8c and d. No significant differences were observed except a littler baseline noise, indicating that our electrode can stably monitor ECG signals despite long-term, continuous wearing. These experimental results indicate that our electrode is wearable for more than 1 week and that this electrode can replace the wet electrode for use in long-term, unsupervised monitoring of personal health.

4. Conclusion

In this paper, we successfully patterned a metal layer on a thin PDMS substrate that was flexible and biocompatible, and we produced a PDMS-based surface electrode for the long-term measurement of ECG signals. Even though the impedance and the motion artifact of the PDMS electrode were somewhat inferior to those of the Ag/AgCl electrode, the PDMS electrode measured ECG signals with comparatively good fidelity. Our PDMS electrode may not replace the conventional Ag/AgCl electrode. However, in special applications such as long-term, unsupervised monitoring, our dry electrodes offer benefits over wet-electrode technology, and these requirements increase rapidly as ubiquitous and mobile health care technology progresses. In addition, we have discovered that our dry electrode has no negative influence on the skin, even after 1 week of continuous wear. This finding reflects a noteworthy advantage of the surface electrode for long-term monitoring.

Acknowledgment

This study was supported by a grant of the Korea Health 21 R&D Project, Ministry of Health & Welfare, Republic of Korea (02-PJ3-PG6-EV10-0001).

References

[1] A. Karilainen, T. Finnberg, T. Uelzen, K. Dembowski, J. Muller, Mobile patient monitoring based on impedance-loaded SAW-sensors, IEEE Trans. Ultrason. Ferroelectr. Freq. Control 51 (2004) 1464–1469.

[2] Breakthrough in Long-term Cardiac Ambulatory Monitoring—Finally a Dry Electrode for Long-term Monitoring, Business Briefing: European Cardiology, (2005) 1–3.

[3] R. Wijesiriwardana, K. Mitcham, T. Dias, Fibre-meshed transducers based real time wearable physiological information monitoring system, in: Proceedings of the Eighth International Symposium on Wearable Computers (ISWC'04), 2004, pp. 40–47.

[4] M. Braecklein, C. Moor, I. Tchoudovski, A. Bolz, New system for cardiological home monitoring with integrated alarm function, 2nd OpenECG Workshop (2004) 51–52.

[5] M. Coyle, Ambulatory cardiopulmonary data capture, in: Second Annual International IEEE-EMBS Special Topic Conference on Microtechnology in Medicine & Biology, 2002, pp. 297–300.

[6] J.G. Webster, J.W. Clark, in: J.G. Webster (Ed.), Medical Instrumentation, Willey, New York, 1998.

[7] A. Searle, J. Kirkup, A direct comparison of wet, dry and insulating bioelectric recording electrodes, Physiol. Meas. 21 (2000) 271–283.

[8] D. Prutchi, A. Sagi-Dolev, New technologies for in-flight pasteless bioelectrodes *Aviation*, Space Environ. Med. 64 (1993) 552–556.

[9] G. Ruffini, S. Dunne, E. Farrés, J. Marco-Pallarés, C. Ray, E. Mendoza, R. Silva, C. Grau, A dry electrophysiology electrode using CNT arrays, Sens. Actuators A: Phys. 132 (2006) 34–41.

[10] A. Karilainen, S. Hansen, J. Müller, Dry and capacitive electrodes for long-term ECG-monitoring, Eighth Annual Workshop on Semiconductor Advances for Future Electronics, 2005, pp. 155–161.

[11] A. Folch, M. Toner, Cellular micropatterns on biocompatible materials, Biotechnol. Prog. 14 (1998) 388–392.

[12] S.G. Charati, S.A. Stern, Diffusion of gases in silicone polymers: molecular dynamics simulations, Macromolecules 31 (1998) 5529–5535.

[13] J.Y. Baek, G.H. Kwon, J.Y. Kim, J.H. Cho, S.H. Lee, K. Sun, S.H. Lee, Stable deposition and patterning of metal layers on the PDMS substrate and characterization for the development of the flexible and implantable micro electrode, Solid State Phenomena 124–126 (2007) 165–168.

Biographies

Ju-Yeoul Baek was born in Gyeonggi-do, Korea in 1969. He received the BS, MS and PhD degrees in Physics from Dankook University, Korea, in 1992, 1994 and 1999, respectively. He was researcher in the Agency for Technology and Standards, Ministry of Commerce, Industry and Energy (MOCIE) of Korea, in 2001. From 2001 to 2005, he was a senior researcher in the Intelligence BioMEMS Laboratory at the Dankook University, Korea. He is currently a Research Professor at the Department of Biomedical Engineering at the Korea University, Korea. His current research interests are the development of BioMEMS devices, such as microvalve, micropump, microchannel and the flexible implantable microelectrode for the biomedical applications.

Jin-Hee An received the BS and the MS degrees in Physics from the Dankook University in Korea, in 2004 and 2006, respectively. In 2006, she was an Researcher at Korea Artificial Organ Center and Department of Biomedical Engineering in Korea University. Her current research interests are the microvalve, pH-responsive microvalve, flexible implantable microelectrode.

Jong-Min Choi received the BSc degree from the School of Electrical Engineering, Seoul National University, Seoul, Korea, in 2000. He is currently working towards the PhD degree in interdisciplinary program in Biomedical Engineering at Seoul National University. He joined the Advanced Biometric Research Center in 2001, and he is currently working on biomedical signal processing in the field of otolaryngology.

Kwang-Suk Park (M'78) was born in Seoul, Korea, in 1957. He received the PhD degree from the Department of Electronics Engineering, Seoul National University, Seoul, in 1985. Since 1985, he worked in the field of biomedical engineering, especially for biological signal measurements and processing. Currently, he is running projects on ubiquitous healthcare mainly focusing on nonintrusive monitoring of biological signals. He is currently a Professor at Department of Biomedical Engineering, College of Medicine, Seoul National University. He is also the Director of Advanced Biometric Research Center in Seoul National University.

Sang-Hoon Lee received the BS degree in Electrical Engineering and the MS and PhD degrees in Biomedical Engineering from the Seoul National Univer-

sity in Korea, 1973, 1987 and 1992, respectively. From 1992 to 1998, he was an Instructor and Assistance Professor, and from 1985 to 1992, he was Researcher at the Department of Biomedical Engineering at the Seoul National University Hospital. From 1998 to 2006, he was Professor at the Department of Biomedical Engineering at the Dankook University. He was Visiting Scientist at the Department of Biomedical Engineering at the University of Wisconsin-Madison, in 2002. He is currently Professor at the Department of Biomedical Engineering at the Korea University, in 2006. His current interests are the development of single cell handling and analysis of microfluidic devices and flexible implantable sensor for medical applications.