ACS SENSORS

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Transformable Electrocardiograph Using Robust Liquid–Solid Heteroconnector

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Cite This: ACS Sens. 2021, 6, 212–219 Read Online

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ABSTRACT: In this study, a highly transformable electrocardiograph that can considerably deform the position of stretchable electrodes based on the lead method for diagnosing heart disease was developed; these electrodes exhibited high resistance stability against considerable stretching and multiple stretching. To realize the large deformable functionality of the electrodes of a system, liquid metal electrodes and a heteroconnector composed of a liquid metal paste and carbon-based conductive rubber were employed. The developed device can achieve a 200% strain with only 6% resistance change and a high stability of resistances after the 100-time stretching test. In addition, the study demonstrated electrocardiograms in different lead methods of adult and child using the same device. The proposed combination of large deformable electrodes with high electric stability and a robust heteroconnector is an important technology, and it presents a considerable advancement in the application of stretchable electronic systems.

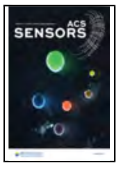
KEYWORDS: *wearable ECG, liquid metal, electric connector, stretchable electronics, stretchable electrodes*

Electrocardiographic measurements are essential for diagnosing heart disease. A standard 12-lead electrocardiogram (ECG) that uses 10 electrodes attached to the body surface to simultaneously measure 12 different ECGs is widely used.[1,2] Further, there are methods such as using a Holter monitor electrocardiograph that can be employed to measure ECG over a long time.[3] Long-term measurement is useful when attempting to measure ECG during heart attacks, for which the time of occurrence is difficult to predict, such as during a stroke. A combination of these short- and long-term measurements is used for diagnosis, that is, continuous electrocardiograph measurement is performed after diagnosing the type of disease with a standard 12-lead ECG.

When measuring ECG for diagnosing heart disease, it is necessary to perform the measurement using the appropriate electrode position based on the purpose of the measurement, that is, identifying the possibility of a specific disease. Based on the position of the electrodes, specific portions of the heart can be evaluated in detail from each waveform. A measurement method that uses the appropriate electrode positions is called the lead method. To use the ECG for diagnosing heart disease, it is necessary to measure ECG using an appropriate lead method.

To date, various types of wearable ECG devices have been developed, such as devices that are miniaturized and integrated with electrodes and attached to the chest,[4,5] armband type devices,[6,7] and clothing type devices.[8] Measurements performed using a wearable ECG can reduce the burden on the patient because of smaller and thinner devices that enable measurement at any locations. Thus far, advanced wearable ECG devices include devices made of flexible[9,10] and stretchable materials.[4,11,12] To add stretchability to the electrodes and sensors, electrodes that have a stretchable structure, such as a meander structure[13−15] or a wrinkle structure,[16−19] and a conductive polymer material into which a conductive material, such as silver flake, can be embedded have been designed on large stretching substrates. The stretch rate of these electrodes, while maintaining the repeatability and stability of the electrical characteristics, is currently limited to approximately 20%,[20] which is sufficient to achieve a suitable interface between the device and the human and to follow stretching and deformation of the human skin. Even with a conventional stretchable wearable electrocardiograph that does not support the conventional lead method,[21,22] it is possible to measure ECG. However, because it is necessary to diagnose

Received: October 12, 2020
Accepted: December 18, 2020
Published: January 4, 2021



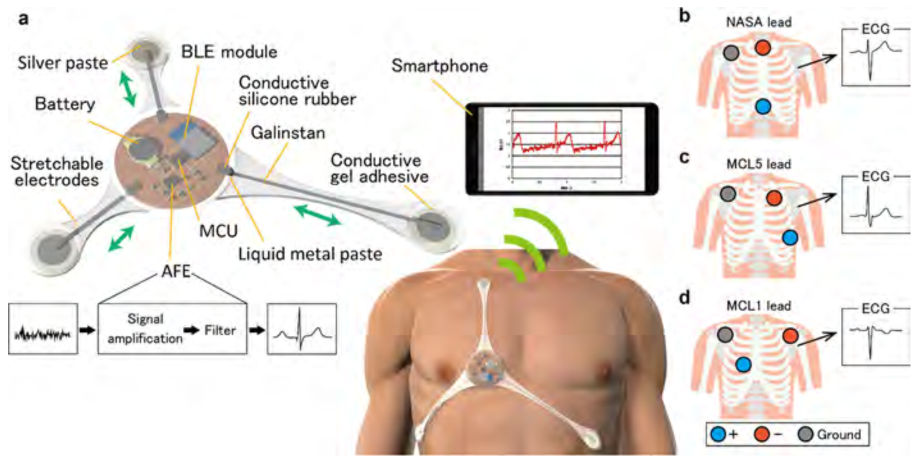


Figure 1. Schematic of the transformable ECG device. (a) Overview of the device. The flexible substrate including AFE, MCU, and BLE for the system part is arranged at the center of the device. The stretchable electrodes are assembled with the system in three directions. A liquid metal paste and conductive silicone rubber form a heteroconnector for linking the stretchable electrode and the substrate. Silver paste covers the edge of the liquid metal electrodes and it binds the liquid metal and conductive gel adhesive. In the measurements, the device is fixed to the body surface using an adhesive made of gel. Electrode position and representative ECG of (b) NASA lead (c), MCL5 lead (d), and MCL1 lead.

heart disease by looking at the waveform of each lead method, the results measured by such stretchable electrocardiographs can only be used for fitness purposes such as calculating the heart rate. For actual medical practices, a wearable electrocardiograph device that can measure ECG continuously based on a specific lead method is required. Therefore, a large deformation and a high degree of freedom for deforming the electrodes are crucial to achieve this goal.

Stretchable electrodes based on liquid metal satisfy the requirement of such a large deformation. The liquid metal electrodes achieve large deformation and maintain a stable resistance simultaneously even if the electrodes are stretched to more than 350% 4500 times.[23] Further, it is possible to suppress the increase in the resistance value during stretching because the liquid metal deforms freely in the flow path. To exploit these advantages, several smart vital sensors, actuators, and systems using liquid metal have been developed.[24] However, the connection between these stretchable electrodes and the system for realizing stand-alone functionality cannot withstand the stress during stretching. Metal wires,[25] snaps,[26,27] and connector elements[28] are often used to connect the stretchable electrode with the system component. However, the method that uses such hard elements in the connector impairs the elasticity of the electrode and device flexibility. These technological problems of robustness and electrical stability may not be applicable to electrodes using liquid metal and other stretchable electrodes that use other conductive materials, such as silver flake.

In this study, we propose a transformable wearable electrocardiograph that possesses highly stretchable electrodes designed according to lead methods and a robust connection between the electrodes and the system. The stretchable electrode is fabricated by injecting liquid metal into a silicone rubber channel. The electrode position can be changed freely by stretching this electrode. A flexible substrate made of polyimide film is used as the substrate of the system part. To realize robust connection, a liquid−solid heteroconnector composed of conductive silicone rubber and liquid metal paste was developed. This connector allows the electrodes to be stretched without compromising the flexibility of the device or impairing electric stability during stretching. In this study, the device was assumed to be used in medical practice. Thus, the purpose of the device was to exhibit resistance stability during the stretching of electrodes while the patients carried on with their normal lives. As a result, this device achieved transformability according to the lead method and the appropriate measurement conditions for each patient in this study.

■ RESULTS AND DISCUSSION

Figure 1a illustrates an overview of the system. The device is composed of a flexible board for the control system and stretchable electrodes. The flexible board is mounted on a hard silicon substrate, polydimethylsiloxane (PDMS), to prevent the stress from being applied to the interface between the PDMS substrate and the system board during stretching. The system performs processes and communicates signals, and it includes a coin battery (CR1220) for stand-alone functionality. The signals are transferred to a smartphone or PC via a Bluetooth low energy (BLE) module. The stretchable electrode is fabricated by injecting Galinstan into a channel made of soft silicone rubber, (Ecoflex 00-20). The positions of the electrodes can be changed freely by extending them. The device achieves appropriate positioning of the electrodes to obtain precise ECG waveforms based on the physical size of the patients and the lead methods, such as NASA, MCL5, and MCL1, as shown in Figure 1b−d. The robust heterointerconnect between the system board and highly stretchable electrodes is crucial to achieve this large transformability of the device by stretching the electrodes. Liquid metal is an ideal material for fabricating large deformable electrodes because it exhibits high structural and electrical stabilities under deformation. However, the liquid metal exhibits considerably low wettability on different metals and polymers. In addition, because of the high fluidity of the liquid metal, it is difficult to form a bridge between hard and soft substrates, while maintaining a stable electric property during stretching. Therefore, the heteroconnector is fabricated using a conductive polymer composed of carbon nanoparticles and multiwall carbon nanotubes (MWCNTs) and liquid metal paste (liquid metal containing Ni microparticles) (Figure 1a). The conductive polymer realizes robust interconnection

https://dx.doi.org/10.1021/acssensors.0c02135
ACS Sens. 2021, 6, 212−219

between the large stretchable electrodes and the system. Simultaneously, the liquid metal paste achieves appropriate wettability on conductive polymers and electrical stability after multiple instances of stretching. Eventually, the device can measure stable and precise ECG under large deformation based on the physical size and lead method.

Figure 2a demonstrates the fabrication process of the stretchable electrodes using liquid metal. Because the electrodes require contact parts with the body to measure the ECG, the electrodes were composed of two layers: a silicone membrane and electrode-shaped silicone mold fabricated using a three-dimensional (3D) mold. As shown in Figure 2a, the top layer was formed using Ecoflex 00-20 (i), and the uncured Ecoflex 00-20 was coated on this layer for adhesion between layers (ii). In the bottom layer, the PDMS was first cured in the 3D mold (iii). Next, the large stretchable part was cut and removed from the mold (iv), and then Ecoflex 00-20 was poured and cured on this part to construct the hybrid substrate with soft and hard silicones. After fabricating each layer, the two layers was adhered to each other (vii). Galinstan was injected into the microchannels (viii). In the end part that contacts the body, silver paste was used to cover the outlet of the microchannel. Figure 2b shows a high-magnified image of the fabricated electrode. This electrode can stretch to 200% (Figure 2b) at the least. The maximum stretching percentage while maintaining electrical stability was found to be 350%, as shown in Figure S1. As shown in Figure 2c, the change in resistance at 200% stretching was small (6.4%). Even if 200% stretching was repeated 100 times, the electrodes exhibited high electrical stability.

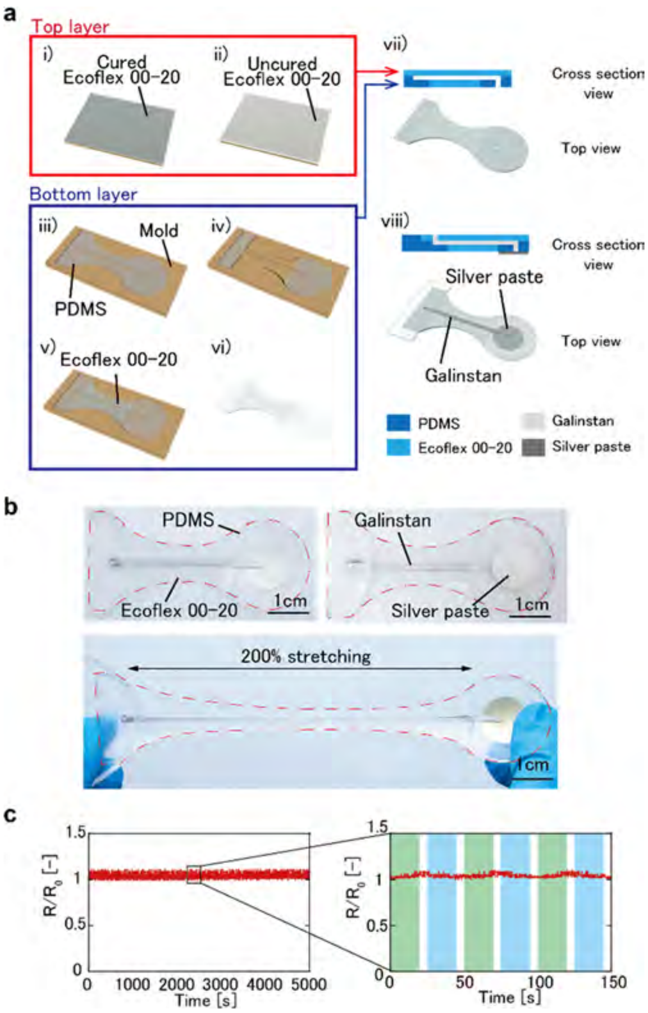


Figure 2. Fabrication method and characteristics of stretchable electrodes for an ECG device. (a) Fabrication process of the electrodes. After preparing the upper layer (i,ii) and the bottom layer (iii–vi), they are bonded by Ecoflex-0020 (vii). After cutting holes at both ends of channel, Galinstan is injected. Silver pastes are applied to the measuring part. (b) Photograph of the fabricated electrodes. The electrodes can be stretched to at least 200%. (c) Repeatability test of electrode stretching. The resistance was stable after the device was stretched 100 times to 200% elongation.

The substrate was constructed using soft and hard silicones. The deformation of the contact part consisting of hard silicone was significantly suppressed to less than 5% as can be seen from the Euler–Armandie strain in Figure S2. This is why the resistance change primarily may occur in the soft part composed of Ecoflex 00-20 during the 200% stretching. As shown in Figure S3, while the resistance value of liquid metal wiring is 0.02 Ω, silver paste and gel have resistance values of 0.03 and 0.01 Ω, respectively. Furthermore, the contact resistance between the liquid metal wiring and the silver paste is 0.12 Ω, and the contact resistance between the silver paste and the gel is 0.35 Ω (Figure S3). Based on the result in Figures S3 and S4, the resistance change of only liquid metal was 7.5% of the total resistance of the electrode because these parts are hardly deformed, and only the resistance value of the liquid metal wiring changes while the electrodes are stretched. However, the resistance of the liquid–solid connector, which does not deform while stretching, is more than 1000 times higher than the total resistance of the electrodes. Therefore, the deformation of the electrode did not affect the functionality of the system during stretching. When a tensile test was performed with the same length of wiring with silver paste as shown in Figure S4, it cracked before stretching to 200%. The normalized resistance (R/R_0) was 35.6 when the stretchable electrode was stretched by 180%.

Electrodes using liquid metal or its composites can realize ultralarge deformations. In other research, liquid metal electrodes were elongated to over 700%.[23] In our study, a maximum elongation of 350% was achieved. Considering the requirements of the device, we believe that this maximum rate of elongation is sufficient. In general, the electrical stability of liquid metal is considerably higher than that of silver paste. As shown in Figure 2, there is no significant difference after elongating up to 200% for 100 times in this study. Regarding the silver paste, the resistance of electrodes increases significantly compared to that for the original electrodes during stretching. Using these advantages, the liquid metal electrodes for the ECG measurement were demonstrated by the Dickey group in North Carolina State University.[29] They realized ECG measurements with extremely low S/N by modifying ion gels between the electrode and the body. In this study, the device was required to improve adhesion between the electrodes and the body. Therefore, adhesive conductive gels were used. In future work, the S/N of the ECG signal may be improved by inserting the ion gel between the liquid metal electrodes and our conductive gel.

The system was assembled on a flexible polyimide film board (Figure 3a). The signals passing through the electrodes were processed by the analog front end (AFE), including amplification and noise removal. The obtained analog signal

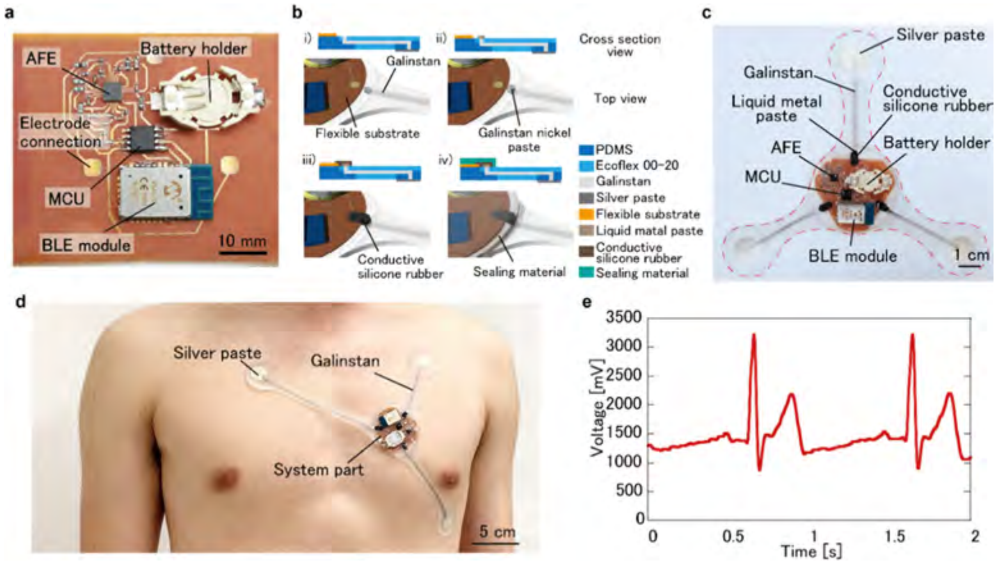


Figure 3. Assembly of a transformable ECG device. (a) Flexible substrate for the system unit. (b) Fabrication process of a heteroconnector composed of liquid metal paste and conductive silicone rubber with CNT and carbon nanoparticle. After injecting Galinstan into the microchannel (i), liquid metal paste (ii) and conductive silicone rubber (iii) were used to form a connector. Eventually, the substrate is covered by silicone rubber adhesive (iv). (c) Photograph of the actual device. (d) The device can be set on the human body with stretching. (e) ECG measured by the developed device.

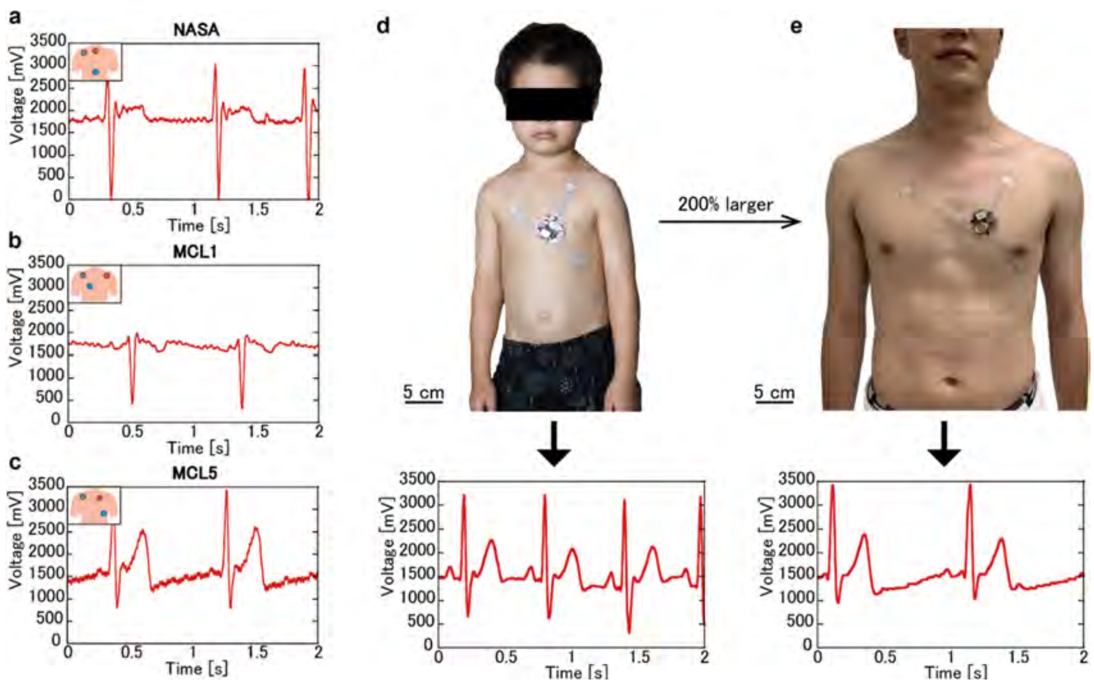


Figure 4. ECG measured by the developed device. (a–c) ECGs measured in NASA (a), MCL1 (b), and MCL5 (c) leads using one device. Each ECG in a different lead can be detected by the same device. (d) The device can detect the ECG for a 6-year-old child. (e) Further, the developed device can measure ECG of an adult whose chest size is two times larger than that of the child by stretching the electrodes.

of electrocardiograph was converted to digital data by the micro controller unit (MCU). The converted data were transmitted to the smartphone via a BLE module; the data were recorded on the smartphone. The ECG was measured using three stretchable electrodes: two electrodes were employed for measurement and the third one was employed for ground. The circuit diagram is shown in Figure S5. Conductive silicone rubber and liquid metal paste were used to connect the electrodes to the substrate (Figure 3b). Conductive silicone rubber was prepared by mixing MWCNT and carbon nanoparticles with silicone rubber.[30] A metal paste[31] prepared by mixing Galinstan and nickel microparticles was used between Galinstan and the conductive silicone rubber to stabilize the connection between them (Figure 3b). The manufacturing of the device was completed using these connectors. Figure 3c demonstrates the fabricated device. Figure 3d presents the state of the device during measurement. Conductive gel adhesive was used between the electrodes and human body to stabilize the position of the device. Figure 3e shows the measured electrocardiographic waveform. In this graph, P, QRS, and T waves, which are important in the diagnosis of heart diseases, were confirmed.

There are several methods for establishing a connection, such as by using a metal wire,[25] conductive tape,[32] conductive adhesive,[33] and connector element;[28] these methods use hard materials for connection. Due to the poor wettability of the liquid metal with the flexible substrate, the wiring breaks during stretching, as shown in Figure S6a. Further, the silver paste breaks due to lack of stretchability, as shown in Figure S6b. As shown in Figure S6c, in the case of a connection with the conductive silicone rubber, the paste stretches similar to the surrounding elastic materials. Therefore, it can be connected to the substrate without impeding the elasticity of the electrode. However, the use of a connector for electrode connection makes a circuit bulky, thereby limiting the circuit design, including the positioning and shape of the electrodes. The conductive silicone rubber connection enables the electrode to connect to a free position in a small space.

In the proposed wearable electrocardiograph, the device was attached to the body with a conductive gel adhesive during measurement. When the electrode was stretched, a tensile force was generated. As shown in Figure S7, the tensile force increased with respect to the strain, which is the same as the results in other research.[34] The tensile force was approximately 1 N at a 200% stretching. The electrode could be attached with a gel until the tensile force reached 3.4 N at a 330% stretch. This adhesive strength is sufficient to hold the device when the subject moves (Video S2). Using this conductive gel adhesive, the contact impedance could be reduced simultaneously as the device was fixed, thereby reducing the hum noise. The conductive gel adhesive could be attached/detached multiple times to adjust the position of the electrode (Figure S8). The tensile force decreased according to multiple cycles. After 20 repetitions, the tensile force, when the conductive gel adhesive detached from the skin, reduced to approximately 1.7 N, as shown in Figure S9. However, the signals were stable after the multiple cycles, as shown in Figure S8. Even if the adhesive strength of the gel decreases after repeated attachment/detachment, the adhesive force would recover by replacing only the gel, and the device could be used repeatedly.

In this device, the electrodes can be placed within a range of 130 mm with the electrode not extended and 270 mm with the electrode extended to its maximum. This range is sufficient to measure the ECG arbitrarily on the chest of adults and children. Furthermore, this device can be used not only for adults and children but also for infants by utilizing smaller electric components and stretchable materials for fabricating electrodes that possess a lower Young's modulus.

Using the developed device, measurement with different leads was performed as shown in Figure 4a−c. NASA lead, MCL1 lead, and MCL5 lead among the lead methods are used commonly for continuous electrocardiograph measurement. P, QRS, and T waves could be confirmed in all lead methods. In addition, difference in the waveform depending on the lead method was observed. During diagnosis, the NASA, MCL5, and MCL1 leads are used for arrhythmia, myocardial infarction and angina, and premature atrial contraction and atrial fibrillation, respectively. The S/N of the signal measured by the wearable electrocardiograph is 25−30 dB.[35,36] In this device, the signals of the NASA, MCL1, and MCL5 leads were 31, 41, and, 26 dB, respectively. Compared with other studies,[35,36] detection of ECG using our devices was feasible.

Further, we measured the MCL5 lead in two subjects with different physiques and compared their waveforms (Figure 4d,e). There was no significant difference between the measurement results for the 6-year-old child and the adult. Using the developed device, it was possible to perform measurements using appropriate electrode positions without being affected by differences in body size (Figure 4d,e). Moreover, long-term measurements using this device were performed, as shown in Figure S10. Adequate ECG was observed 6 h after the start of the measurement. Video S1 shows that it is also possible to measure ECG while performing daily activities, such as standing up and sitting down from a chair and climbing stairs. In addition, the device could measure ECG by different lead methods, as shown in the video.

Numerous conventional wearable electrocardiographs have fixed electrode positions.[10,37] In various cases, two electrodes are placed on the chest several centimeters apart. The electrode positions of these electrocardiographs do not correspond to the lead method. The signal is small because the electrodes are close to each other. Such a device can be equipped with fitness application devices, in which the heart rate is calculated from the interval between QRS waves, or with devices used by healthy people for healthcare purposes. However, it is difficult to diagnose specific heart disease symptoms from the waveform. In addition, it is necessary to change the device design itself based on the lead method employed because the electrode shape is almost fixed.[35] Using the developed wearable ECG, which has highly stretchable electrodes, it is not necessary to change the device design for different lead methods. Further, by stretching the electrodes, the electrode position can be altered according to an arbitrary lead method, and the electrode position can also be adjusted according to the difference in the physical constitution of the individual.

To observe changes in the waveform when the electrode position is changed, the waveform is measured by moving one of the measurement electrodes (Figure 5a−d). Figure 5a shows the result of the measurement with the positive measurement electrode approaching the side of the negative measurement electrode from the NASA lead of Figure 5b. The Q wave in Figure 5a was lower than that in Figure 5b. Further, Figure 5c,d presents the measurement results when the positive measuring electrode was moved away from the negative measuring electrode. Figure 5e shows the result of superimposing Figure 5a,b,d on a single diagram. By changing the position of the electrode, the direction of each wave, from which the electrical signal in the heart is observed, changes. Therefore, the obtained ECG waveform also changes.

Accordingly, the obtained electrocardiographic waveform changes when the electrode position shifts. The influence of the displacement of the electrode position on the ECG is not a significant problem in the case of detection of a normal ECG. However, for diagnosing or monitoring specific heart diseases, a displacement of more than 2 cm in the electrode position can have a substantial effect.[38,39] The fine adjustment of the electrode position is crucial to obtain the required ECG accurately. Because the electrode positions can be changed, unlike in a conventional wearable electrocardiograph,[37] it may be possible to measure the ECG effectively for diagnosis. In future work, by improving the stability of the ECG signal, it will be possible to perform ECG monitoring during intense movements such as in sports.

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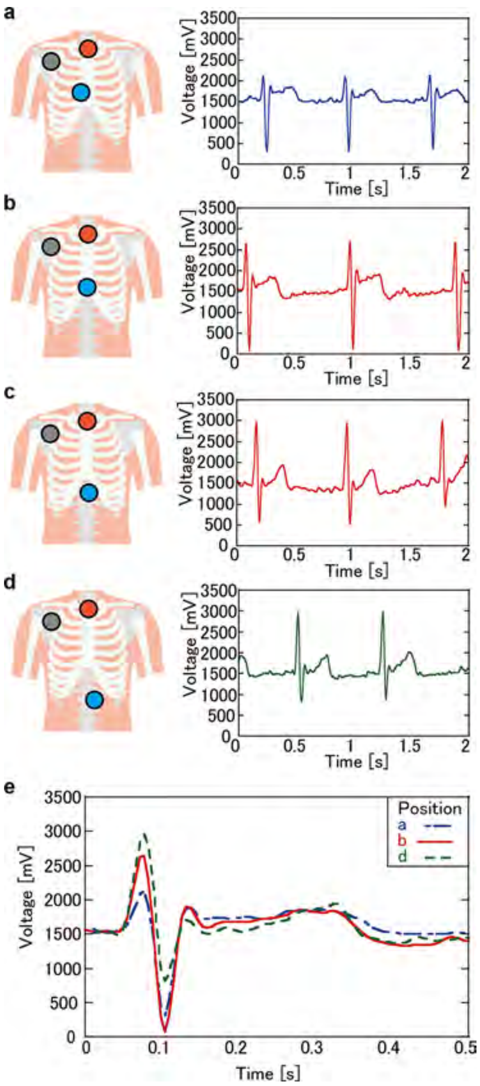

Figure 5. Changes in ECG based on the electrode location. (a−d) ECGs were measured in different locations of the negative electrode. The shape of the ECG was different based on the distance between the negative, positive, and ground electrodes. (e) The result of the multiple ECG signals measured in different measurement positions.

■ CONCLUSIONS

We designed a wearable electrocardiograph with a stretchable electrode using a liquid metal flow path. The wearable electrocardiograph could change the lead method based on the purpose of measurement and adjust the electrode position according to the difference in body size using highly expandable electrodes. To achieve this functionality, a heteroconnecter composed of conductive silicone rubber and liquid metal paste was developed. The electrode can stretch by 350%, and the change in the resistance value during the stretch can be kept relatively small. Our heteroconnector can withstand stresses up to 3.4 N. This robustness of stress applied to electrodes might be in high demand not only for stretchable electronics but also for flexible electronics based on flexible polyimide or polyethylene terephthalate films. This is because the interconnection of the flexible electronics between the electrodes and system is also exposed to the stress because of the difference in the Young's modulus. The drawback of our connector is the high resistance of the connector compared with that of the electrodes. In the systems of ECG or electromyogram, high resistors are mounted between electrodes and systems. Therefore, our connector is useful. However, in terms of devices that detect small signals such as pulse oximeters, no high resistors exist between the electrodes and system. In future work, we will aim to improve the conductivity of the conductive rubber with high conductivity and stability of deformation. It is possible to improve the conductivity by establishing liquid metal paths inside the rubber. Thus, our device that can measure ECG using an appropriate lead method based on the disease can aid in increasing the versatility of electrocardiographic monitoring with a wearable electrocardiograph. In addition, it can become a multivital monitoring device by combining it with sensors for measuring vital signs, such as blood oxygen saturation and body temperature, thereby suggesting its potential applications to other diseases other than cardiac disorders. Therefore, a combination of large deformable electrodes with high electrical stability and a robust heteroconnector present considerable advancements in the application of stretchable electronic systems.

■ EXPERIMENTAL SECTION

Fabrication of Electrodes. For the top layer, Ecoflex 00-20 was spin-coated on an acrylic plate at 200 rpm for 20 s and cured at 70 °C for 20 min. Ecoflex 00-20 was spin-coated again at 1000 rpm for 60 s. For the bottom layer, liquid PDMS (Sylgard 184, Dow Corning) was poured into an acrylic mold for the upper layer and held at 0.08 MPa for 30 min and then cured at a 70 °C for 1 h. The middle part of the PDMS was discarded. Ecoflex 00-20 (Smooth-On) was poured on this middle part and cured at 70 °C for 20 min. After detaching the substrate from the mold, a hole (1.5 mm in diameter) was cut in the shape of an electrode at the end of the flow path (contact side with the human body). The bottom layer was overlaid on the upper layer and cured in an oven for 20 min. A 1.5 mm hole was cut on the circuit side at the end of the flow channel. Liquid PDMS was poured into a mold made using a 3D printer (Form2, Formlabs), defoamed, and cured (70 °C, 1 h in oven). The cured PDMS and the electrodes were bonded together with silicone rubber adhesive (Sil-poxy, Smooth-On). Galinstan was injected into the channel, and silver paste was applied to the part in contact with the human body and cured (70 °C, 1 h in oven).

Fabrication of the ECG Measurement System and Data Transmission System. The circuit diagram of the ECG measurement system is shown in Figure S5. A 180 kΩ protection resistor was incorporated between the electrode and the circuit to prevent strong current from flowing through the human body. The potentials at two points on the body surface were read with electrodes, and the potential difference, that is, the electrocardiographic signal, was obtained using a differential amplifier circuit in AD8232 (Analog Devices). Because the ECG signal is in the range of several millivolts, it was smaller than the resolution of the analog digital converter, and the signal was amplified 1100 times by the differential amplifier and the noninverting amplifier in AD8232. To remove noise caused by body motion and hum noise caused by commercial power, a 50 Hz high-pass filter and a 0.5 Hz low-pass filter were constructed using the operational amplifier in AD8232 and the surrounding resistors and capacitors. ATtiny85 (Microchip Technology) converted the ECG signal to a digital signal at 5.5 ms intervals. The signal was sent to the smartphone via RN4020 (Microchip Technology).

Preparation of Conductive Silicone Rubber. 110 mg of MWCNT and 110 mg of Super P carbon black were mixed with 5 ml of chloroform at 600 rpm for 30 min and sonicated for 30 min. Ecoflex 00-20 (A: 2 g, B: 0.4 g) was added into the solution. This conductive silicone rubber was cured by evaporating the solvent chloroform in the draft and then heating it in an oven at 70 °C for 30 min.

https://dx.doi.org/10.1021/acssensors.0c02135
ACS Sens. 2021, 6, 212−219

Preparation of Modified Liquid Metal. 30 g of Galinstan and 4 wt % of nickel microparticles were mixed by an ultrasonic probe at 2000 rpm for 6 min. Six cycles of sonication were performed twice. One cycle consisted of 2 s ON and 4 s OFF.

System Integration. Using the silicone rubber adhesive, the flexible substrate of the system circuit was bonded to the PDMS substrate for the central part of the device. The modified liquid metal was applied to the channel outlet into which Galinstan was injected. The conductive silicone rubber covered the modified liquid metal and was cured in an oven after volatilizing the chloroform contained as a solvent. The device fabrication was completed by protecting the entire circuit using a silicone sealing material.

ECG Measurement. Three electrodes (two for measuring potential difference and one for grounding the human body) were attached to the body surface using 2268 conductive adhesive (3M Japan Limited) based on the specific lead method. ECG was measured using three types of lead methods: NASA, MCL5, and MCL1. In addition, the ECG was measured with MCL5 lead in two subjects with different physiques. The signals were transferred to a smartphone or PC and recorded.

Electrode Tensile Test. Using a uniaxial stage, the electrode was stretched up to 200% and returned to the original position. The stage speed of the movement was set at 1 mm/s. The strain generated in the electrodes at this time was analyzed using MATLAB.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acssensors.0c02135.

Limit of stretching an electrode; strain distribution after image processing during stretching; tensile tests of stretchable electrode; circuit design of the device; connections between substrates and electrodes; evaluation of adhesive strength of conductive gel adhesive; and long-term ECG measurement (PDF)

Possibility of measuring ECG while performing daily activities (MP4)

Adhesive strength of the device when the subject moves (MP4)

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Author Contributions

M.T. collected all data and led all experiments. R.M. made conductive silicone rubber. G.I. created the data processing system. G.I., U.K., and Y.I. helped designing circuit. D.T. and O.F. helped making acrylic mold. F.N made liquid metal paste. F.N. helped in image analysis of strain. Y.O. helped taking photos of the fabricated device. H.O. guided the research. M.T and H.O. wrote the paper.

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

This work was supported by the Japan Science and Technology Agency, PRESTO Grant Number JPMJPR18J2, MIC/SCOPE (number: 181603007), AMED under Grant Number 20hm0102085h0001, and The SATOMI Scholarship Foundation. H.O. acknowledges support from a Grant-in-Aid for Young Scientists (A) and Grant-in-Aid for Challenging Exploratory Research provided by the Japan Society for the Promotion of Science. The conductive gel adhesive was offered from 3M Japan Limited.

■ ABBREVIATIONS

ECG, electrocardiogram; PDMS, polydimethylsiloxane; BLE, Bluetooth low energy; MWCNT, multi-walled carbon nanotube; MCU, micro controller unit; ADC, analog digital converter

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Transformable electrocardiograph using robust liquid-solid hetero-connector

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Table of contents:

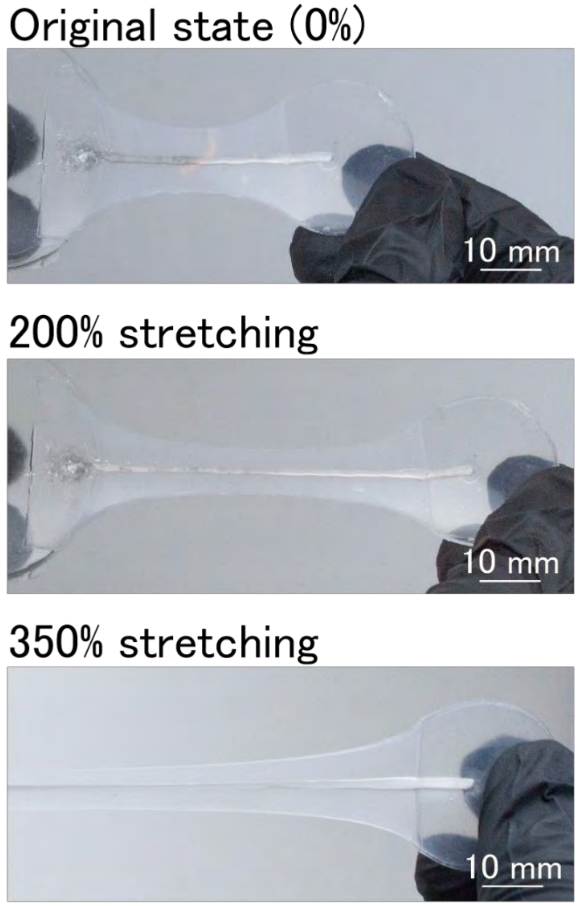

Figure S1 Limit of stretching an electrode.

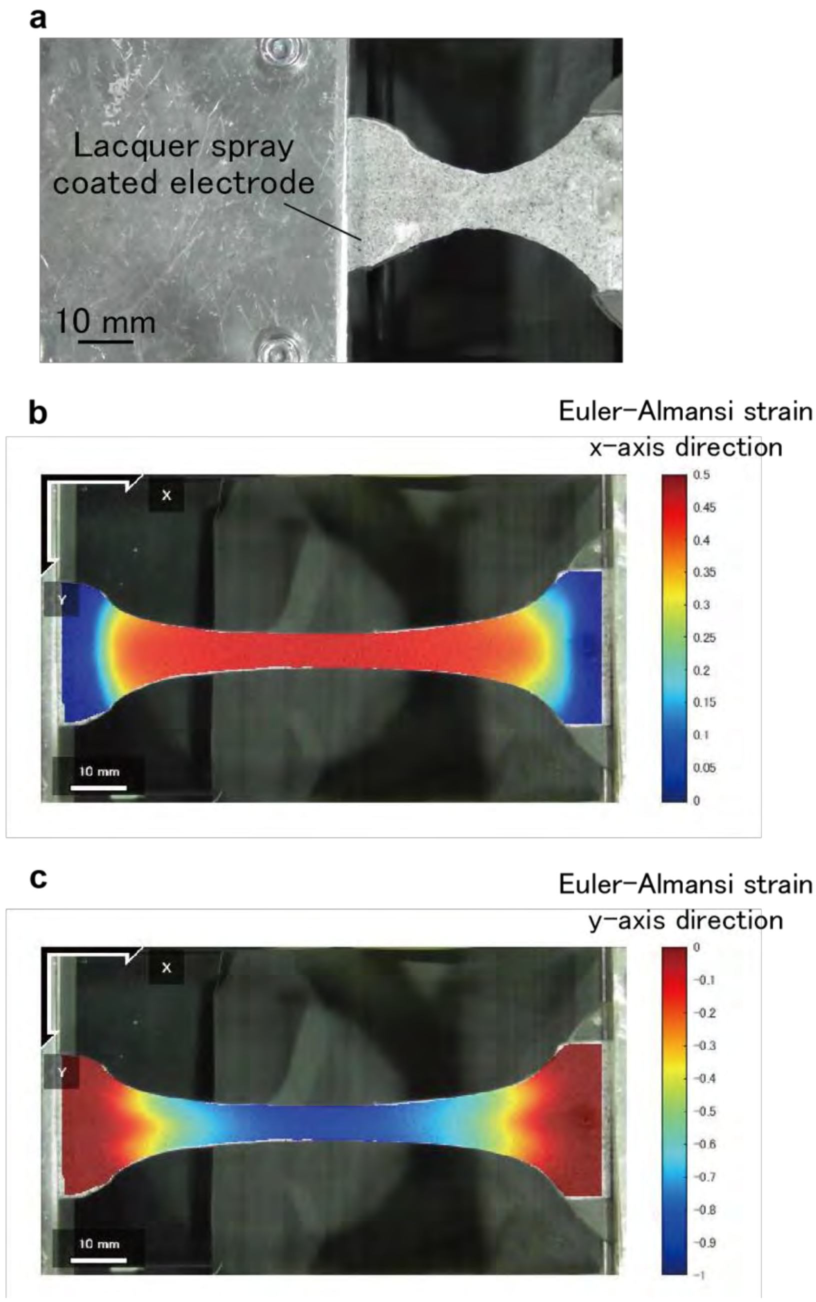

Figure S2. Strain distribution after image processing when the electrode was stretched to 200%. (a) Photograph in strain experimental condition. (b) Distribution of x-axis Euler–Almansi strain at 200% stretching. (c) Distribution of y-axis Euler–Almansi strain at 200% stretching.

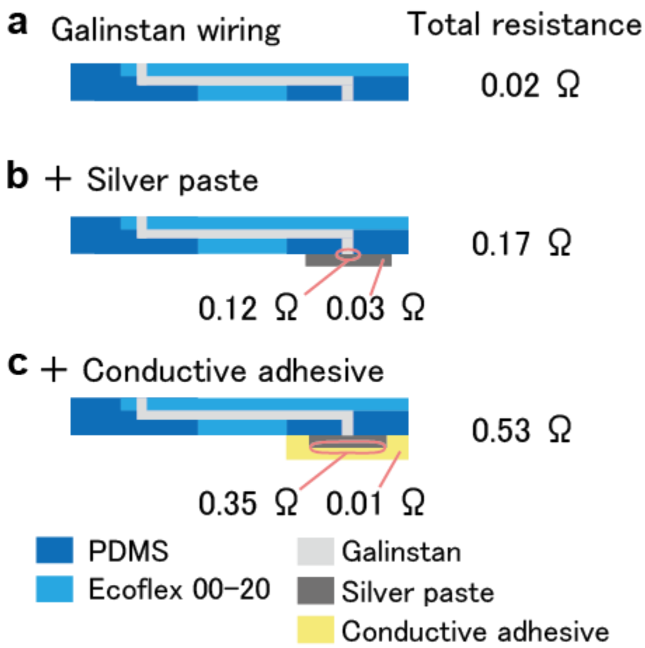

Figure S3. Breakdown of the resistance values of the stretchable electrode. The resistance values of liquid metal only, silver paste coating, and the conductive adhesive attached, and the contact resistance between liquid metal and silver paste and between the silver paste and conductive adhesive.

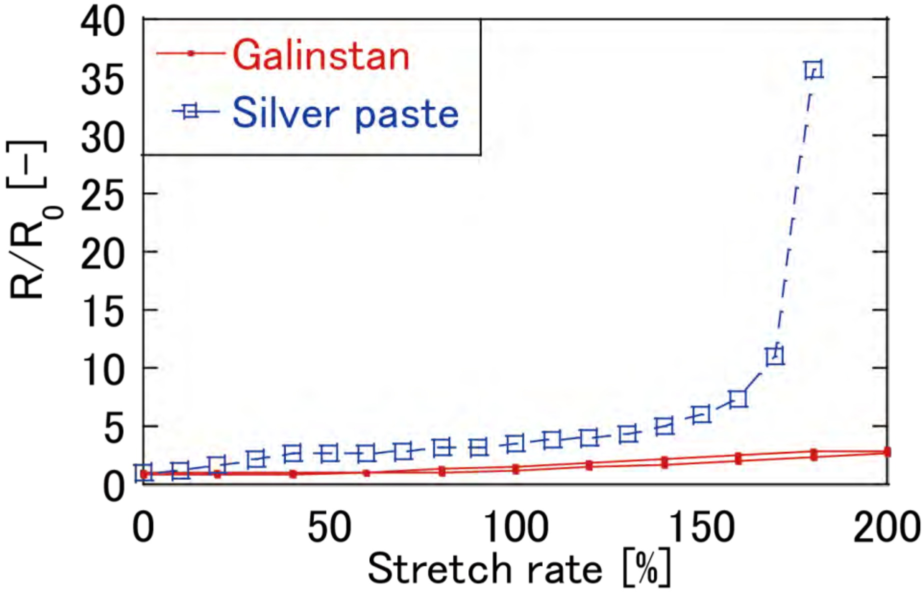

Figure S4. 200% tensile tests using silver paste and Galinstan on a stretchable substrate.

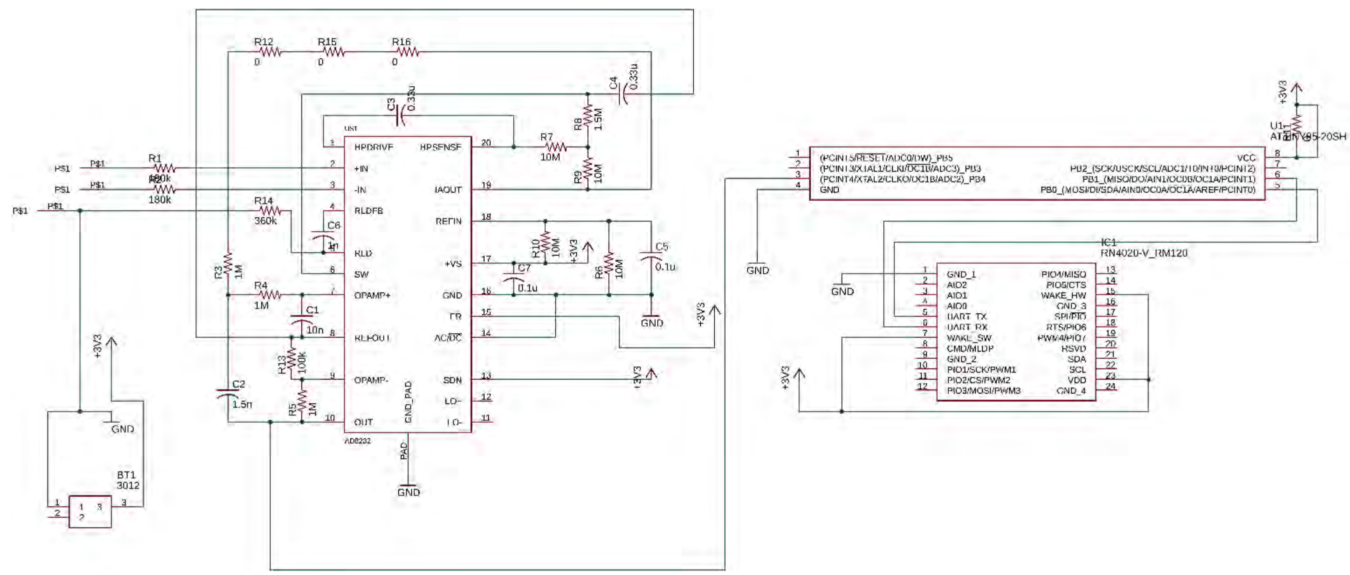

Figure S5. Circuit design of the developed device

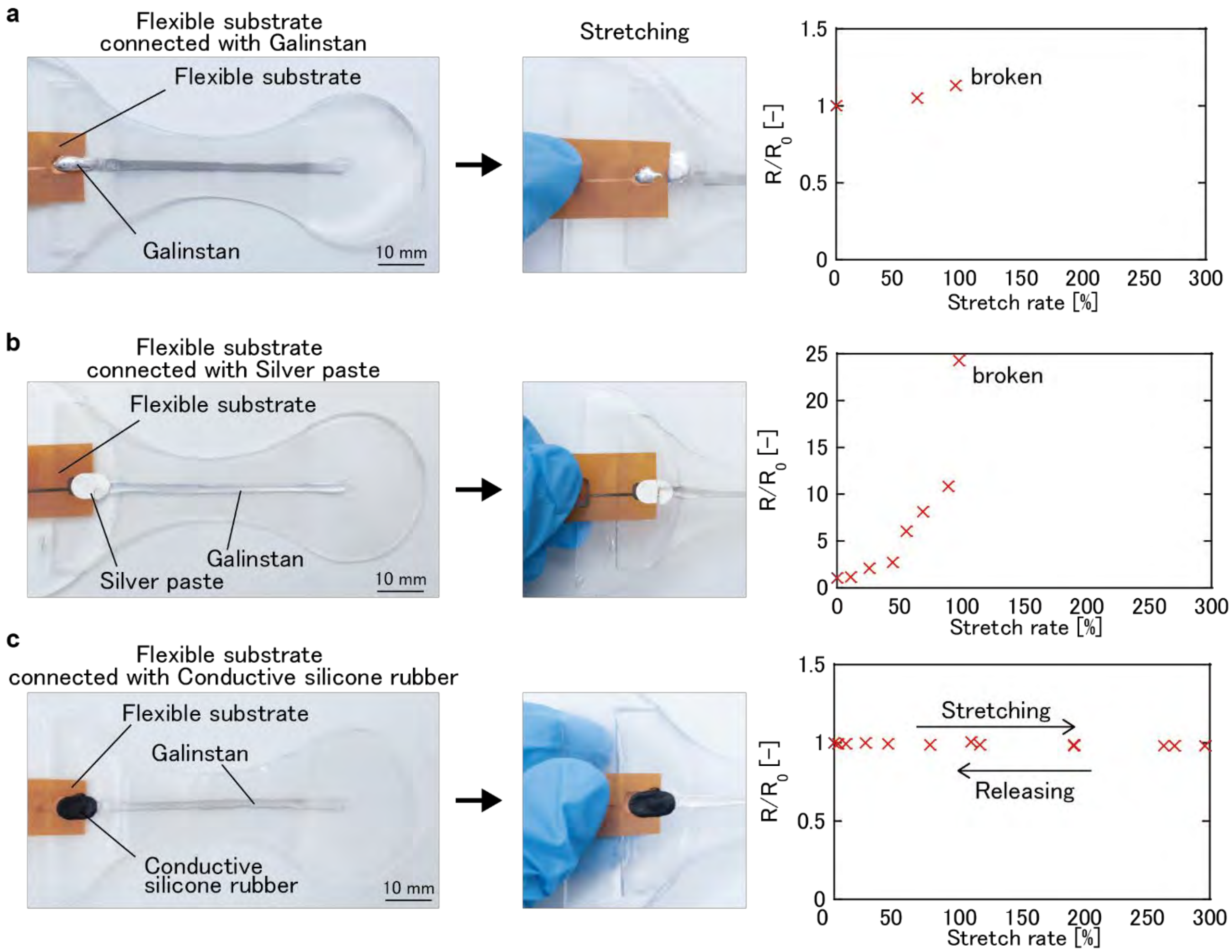

Figure S6. Connections between flexible substrates and electrodes with different combinations. (a, b). When using only liquid metal (a) or silver paste(b), the connection was broken around 100% stretching. In particular, the resistance change was large in the case of the silver paste connection. (c) For the hetero-connector composed of conductive silicone rubber and liquid metal paste, the connection and resistance were stable up to 300%.

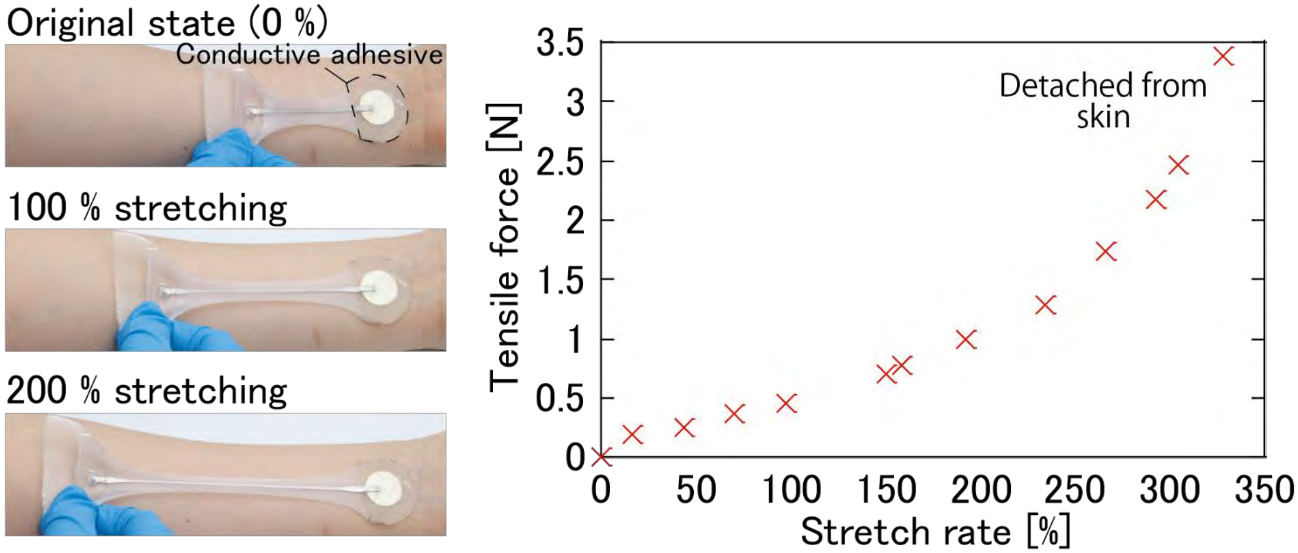

Figure S7. Evaluation of the adhesive strength of the conductive gel adhesive. Even if the electrode was stretched to 200%, the conductive adhesive did not peel off from the body surface. The tensile force when the electrode was stretched increased with strain.

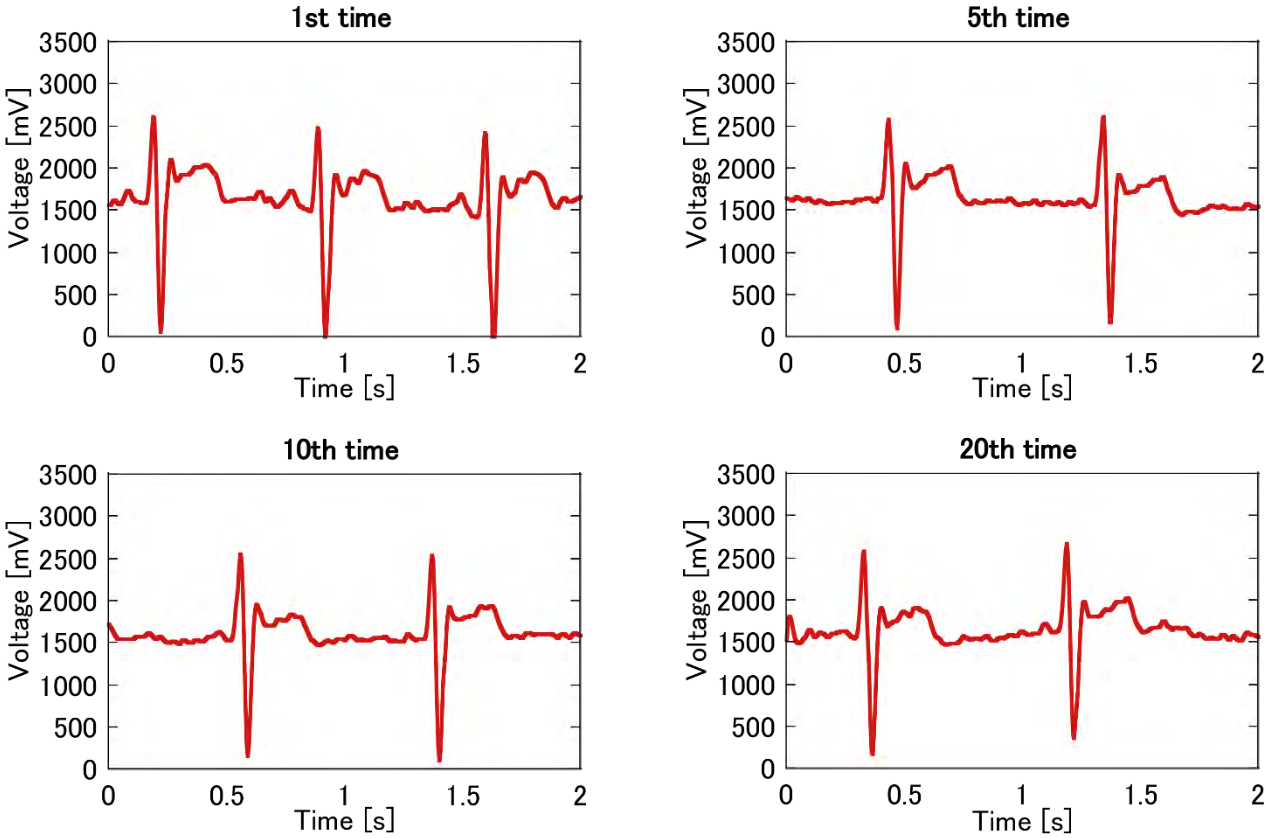

Figure S8. ECG measurements after the electrodes were repeatedly attached/detached

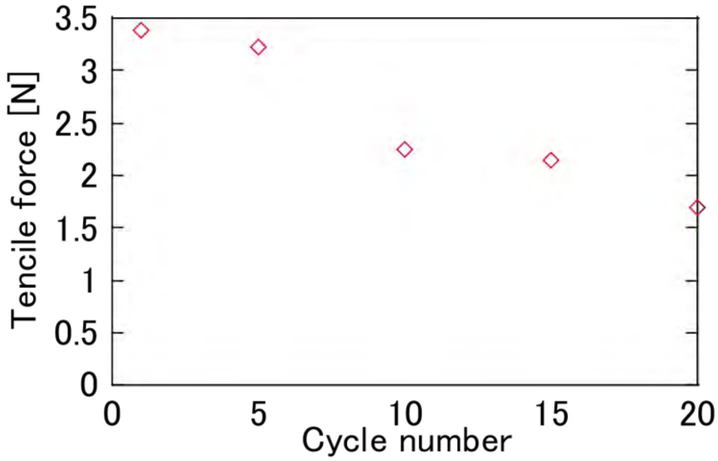

Figure S9. Reduction in the adhesive strength of conductive adhesive with respect to the number of cycles

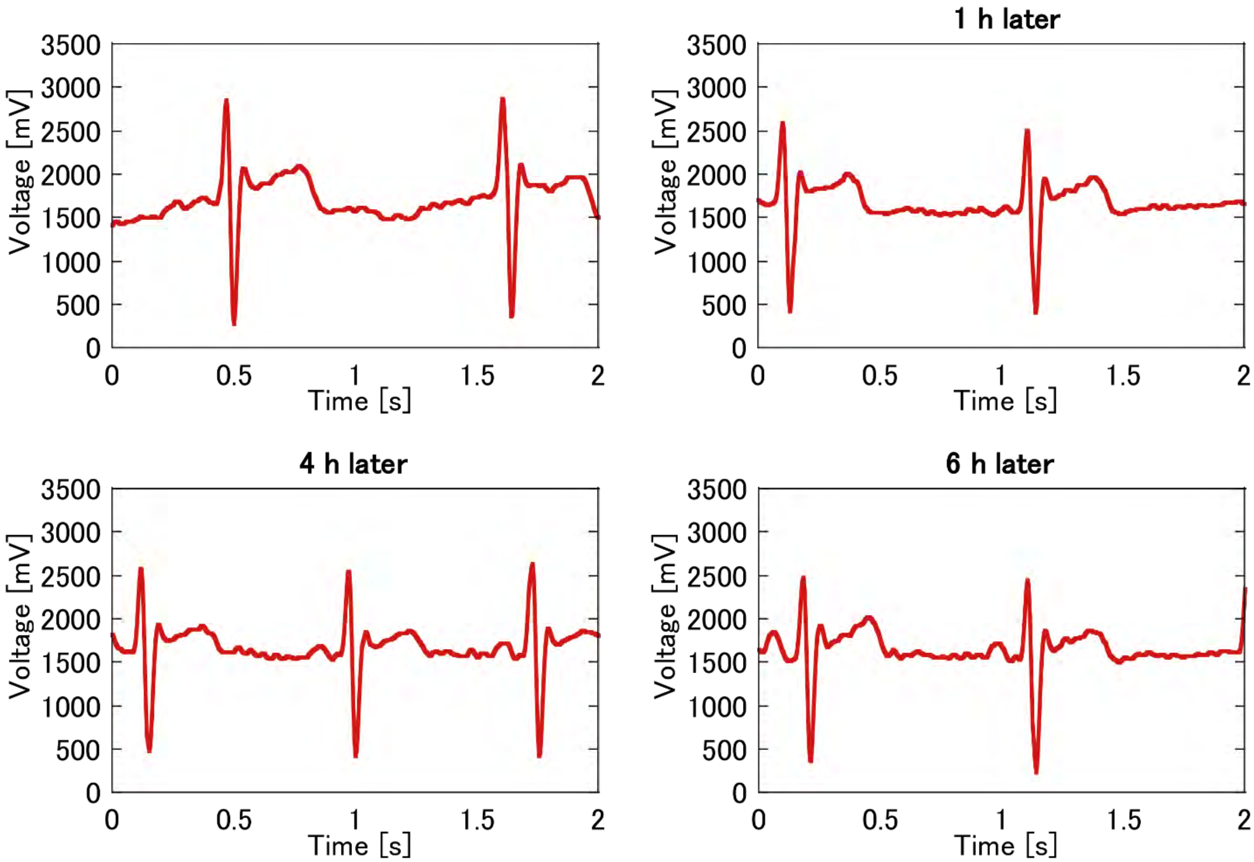

Figure S10. Long-term ECG measurement of the developed device with NASA lead (up to 6 h).

Supplementary video 1. Measurement by ECG device. The measured ECG is displayed on the computer screen. Three actions were performed: 1) standing up and down from the chair; 2) climbing up the stairs; 3) changing the lead method from MCL5 to MCL1;

Supplementary video 2. The adhesion strength of the conductive adhesive was tested. The position of electrode did not shift, and the attached device did not fall off even while running.