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PDMS–ZnO flexible piezoelectric composites for measurement of muscle activity

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Abstract. Measurement of muscle activity is important for muscle health monitoring, biomechanics studies, developing prosthesis, etc. This article describes a flexible piezoelectric composite material as a sensing element for measuring muscle activity. The developed piezoelectric material is a composite of polydimethylsiloxane and zinc oxide, and exists in monolayer and bilayer configurations. To test the piezoelectric properties in bending mode, a composite patch is attached to a cantilever beam setup. Peak sinusoidal voltage generated from the composite material due to the vibrating cantilever is found to be highest (1.5 V) for bilayer configuration with 30 wt% ZnO. For testing in axial mode, the peak output voltage from the material due to an impulse load is maximum (0.9 V) for the monolayer configuration of the composite with 30 wt% ZnO. The sensor consisting of a bilayer composite patch is wrapped around a specific muscle to measure its activity. The change in output voltage from the sensor is measured for increasing load and is then mapped to the corresponding value of elastic modulus of the muscle measured using a durometer. The sensitivity of the muscle activity measurement for biceps brachii and flexor carpi is found to be 3.826 and 1.245 V MPa^{-1}, respectively.

Keywords. Polydimethylsiloxane; zinc oxide; piezoelectric; composite; muscle activity; output voltage.

1. Introduction

Muscle health monitoring is important to prevent the onset of muscular dystrophies or any other muscular disorder [1]. Similarly, estimation of a human's motion intention is one of the key challenges in developing human–robot interaction, such as in smart prosthesis for limb amputees [2–4]. Hence, there is a need to measure and monitor the activity of skeletal muscles in the human body. In the past, various non-invasive sensors measuring muscle pressure [5] and muscle stretching [6] based on the principles of surface electromyography [7], muscle elastography [8], piezoelectric resonance [9], etc. have been reported. However, each of them has several challenges for its practical usage. For example, surface electromyography measurements are extremely sensitive to the changes in impedance of local skin tissue and conductivity between the skin and electrodes [10]. Also, their usage is limited to large clinical settings or research labs. Piezoelectric resonance-based sensors can measure the muscle stiffness accurately, but have limitations due to high driving voltages, thermal drift and variation in thickness of the skin tissue [11]. Some of the limitations confronted by other sensors are a poor spatial resolution, large size of the sensor, the requirement of continuous vertical contact with muscle, etc. Hence, there is a need to develop a sensor that can measure muscle activity in a non-invasive, accurate and reliable manner.

The changes in mechanical properties of skeletal muscles play a significant role in the control of force and motion in humans [12–14]. For instance, the contraction of muscle reduces its length and increases its cross-section area along with the stiffening of muscle. Thus, probing the changes in stiffness of a muscle can help in measuring the muscle activity. Flexible piezoelectric materials, such as polyvinylidene difluoride films, lead zirconate titanate (PZT) nanoribbons on polyimide substrate, PZT fibres in PDMS (polydimethylsiloxane) matrix, PZT particles in epoxy-based polymers, etc., have been used for sensing applications [15]. PDMS is used as a flexible substrate for various sensors as it is an inert, non-toxic and non-flammable elastomer [16]. ZnO (zinc oxide) in its crystalline form of wurtzite structure has a distorted tetrahedral geometry that gives rise to a significant piezoelectric property [17]. Hence, ZnO is preferred as a piezoelectric over PZT, which is toxic and difficult to process. In this study, PDMS–ZnO composite specimens are fabricated and tested for measuring muscle activity. Before the measurement of muscle activity, the composites are characterized

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for their mechanical and piezoelectric properties. Muscle activity of the biceps brachii and flexor carpi muscle on loading the arm is measured using the developed piezoelectric composite material.

2. Experimental

The composite samples of PDMS–ZnO are fabricated in monolayer and bilayer configurations as shown in figure 1a. The constituent materials mentioned in table 1 are obtained for preparing composite samples.

2.1 *Process flow for fabrication of monolayer composites*

The fabrication of monolayer composites starts with: (i) Elastomer base and curing agent, which are uniformly mixed in a ratio of 10:1 by weight, respectively. (ii) Zinc oxide micro-crystalline powder is added according to the required composition (% by weight) of the composite. Specimens are fabricated with the content of ZnO as 25 and 30 wt%. (iii) Then the constituents are stirred till the powder gets uniformly wetted by PDMS solution. (iv) The paste is then poured into a Petri dish on a spin coater set to rotate at 150 rpm for 2 min. (v) Next, the Petri dish is placed inside a microwave oven to cure the sample at 160°C for 10 min. After curing, the sample is allowed to cool down and then gently lifted off from the Petri dish.

2.2 *Process flow for fabrication of bilayer composites*

The process flow for the fabrication of bilayer composites is as follows: (i) Initially, pure PDMS is cured at 150°C for 10 min in the oven. (ii) Then a paste of PDMS and ZnO microcrystalline powder is prepared as explained in the fabrication process of monolayer composites. (iii) The paste is spin-coated on the cured PDMS in a Petri dish at a speed of 150 rpm for 2 min. (iv) The sample is then cured at 160°C for 10 min in an oven. (v) Finally, the sample is slowly lifted off from the Petri dish after it is cooled down to room temperature. Using scanning electron microscopy, we observed a uniform and dense distribution of ZnO particles in PDMS substrate for all the fabricated composite specimens.

2.3 *Testing of mechanical and piezoelectric property of the composites*

Test specimens of size 50 × 50 × 6 mm are cut from the sample for measuring Shore-A hardness as per ASTM D2240 standards. The conical indenter of the tester is pressed on the composite substrate and the hardness of the sample is indicated on the scale of 0–100. The measurement is repeated at six different locations on a specimen and an average value of hardness is recorded. As per literature [18,19], the elastic modulus of these specimens could be calculated using the shore readings as given below:

$$E = \frac{5.49 + 0.75145s}{2.67r(254 - 2.54s)}, \tag{1}$$

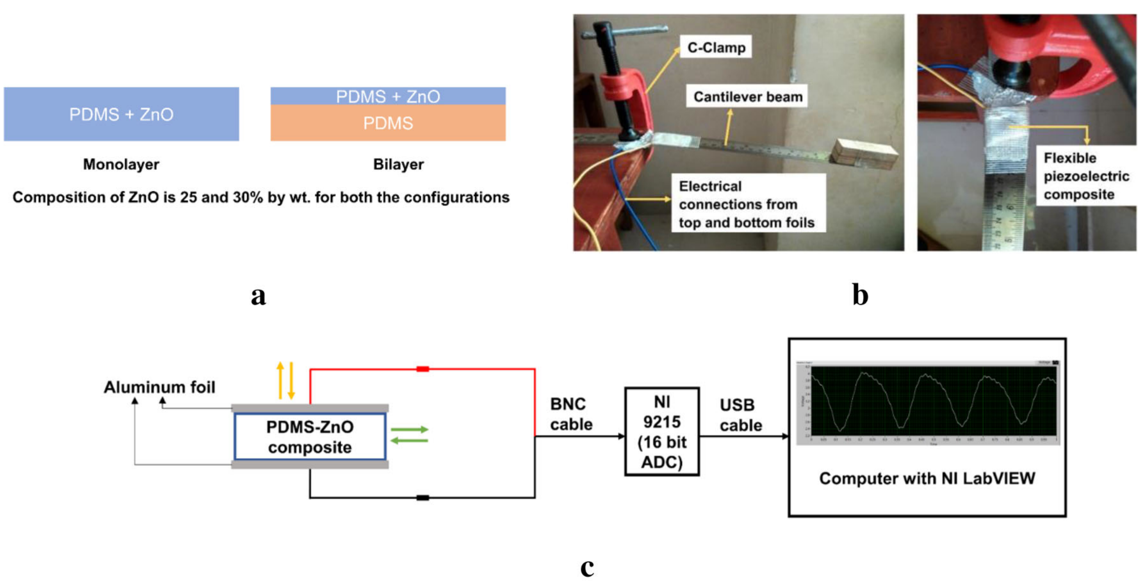

Figure 1. (**a**) Schematic of the monolayer and bilayer configurations of the fabricated composites. (**b**) Side and top views of the setup for testing the piezoelectric properties of the composite in bending mode of operation. (**c**) Schematic of the measurement setup. Green arrows indicate the direction of applied force in bending mode and yellow arrows indicate the direction of applied force in the axial mode of operation.

Table 1. List of constituent materials obtained for preparing the composite samples.

Materials	Trade name	Supplier
PDMS	Sylgard 184 silicon/ elastomer	Dow Chemical/ Company
ZnO	Zinc oxide	Merck

where E is the elastic modulus of the sample, s is the shore hardness of the sample and r is the mean radius (in cm) of the indenter, which is equal to 0.625 mm.

The piezoelectric property of the developed composite specimens in the bending mode is tested in an in-house made setup, as shown in figure 1b. It consists of a stainless steel cantilever beam of length 250 mm and a mass of about 0.05 kg. Specimen with dimensions as $20 \times 25 \times 2$ mm is cut along with aluminium electrodes and then attached to the fixed end of the beam. Leads from the electrodes are connected to a BNC cable that is finally connected to the analog input channel of the NI9215 module, as shown in figure 1c. A mass attached to the free end is pulled down to a fixed height and then released. The sinusoidal voltage output from the piezoelectric composite is recorded in LabVIEW. Fast Fourier transform of the output voltage is taken in real-time, and peak voltage and the resonance frequency of the system are noted for each specimen. Fast Fourier transform of the voltage signal shows a peak close to 4 Hz. The natural frequency of the system calculated from first principles is 4 Hz, thus validating the peak voltage at the resonant frequency.

To test the piezoelectric properties of the composite in axial mode, the composite specimen is first placed between two aluminium electrodes and then placed on a wooden substrate. Now, a fixed impulse load is applied to the composite. The peak voltage generated from the composite due to the impulse is recorded for each specimen.

2.4 *Measurement of Muscle Activity*

Before measuring the muscle activity using the piezoelectric composite, change in stiffness of muscles during a common activity like tightening and relaxing of the fist is first considered. So, the elastic modulus of five different muscles of a subject for relaxed and tightened fist conditions is measured using a durometer (Shore-A). These five muscles are namely: flexor carpi, brachioradialis, extensor carpi ulnaris, biceps brachii and triceps brachii. The location of these muscles in human arm can be found elsewhere [20].

The experiment performed to demonstrate the use of the developed sensor for muscle activity measurement is as follows. First, the elastic modulus of biceps brachii and flexor carpi muscles are measured using a durometer (Shore-A hardness tester) when the subject is holding a weight of 0, 2, 4, 6, 8 and 10 kg, respectively. The arm is maintained parallel to the ground with a weight lifted. Now, a sensor patch consisting of bilayer composite is wrapped around the muscle (figure 2). The change in output voltage from the sensor is recorded when the user lifts the above-mentioned weights. The muscle activity measurements on biceps brachii and flexor carpi are done separately such that the sensor is wrapped only on one of these muscles at a time. An analytical model to calculate the force exerted on the sensor patch due to the muscle pressure/stiffness is given in the supplementary information.

3. Results and discussion

The hardness and elastic modulus of pure PDMS and PDMS–ZnO composite specimens are shown in figure 3a and b, respectively. The hardness and elastic modulus of composites increase with the addition of 25 and 30 wt% ZnO to PDMS.

First, the output voltage measured with only PDMS film was around 0.1 V, which is a measurement artefact. Now, as PDMS is a dielectric material, it can be expected that the

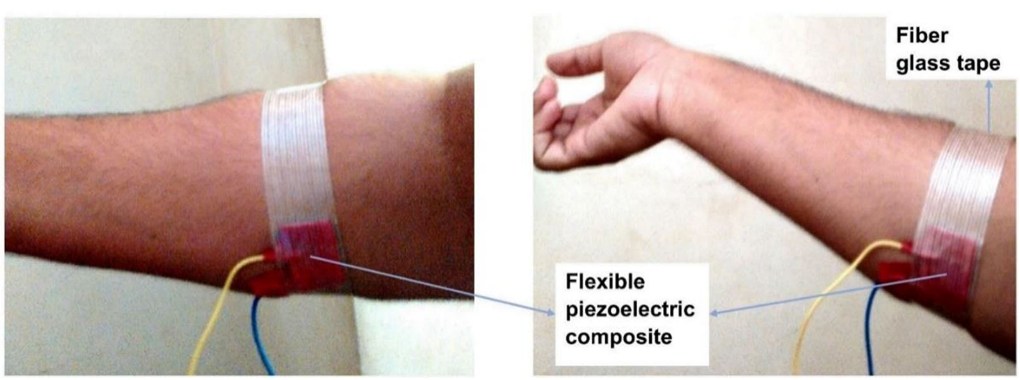

Figure 2. Flexible piezoelectric composite specimen wrapped around the flexor carpi muscle for muscle activity measurement.

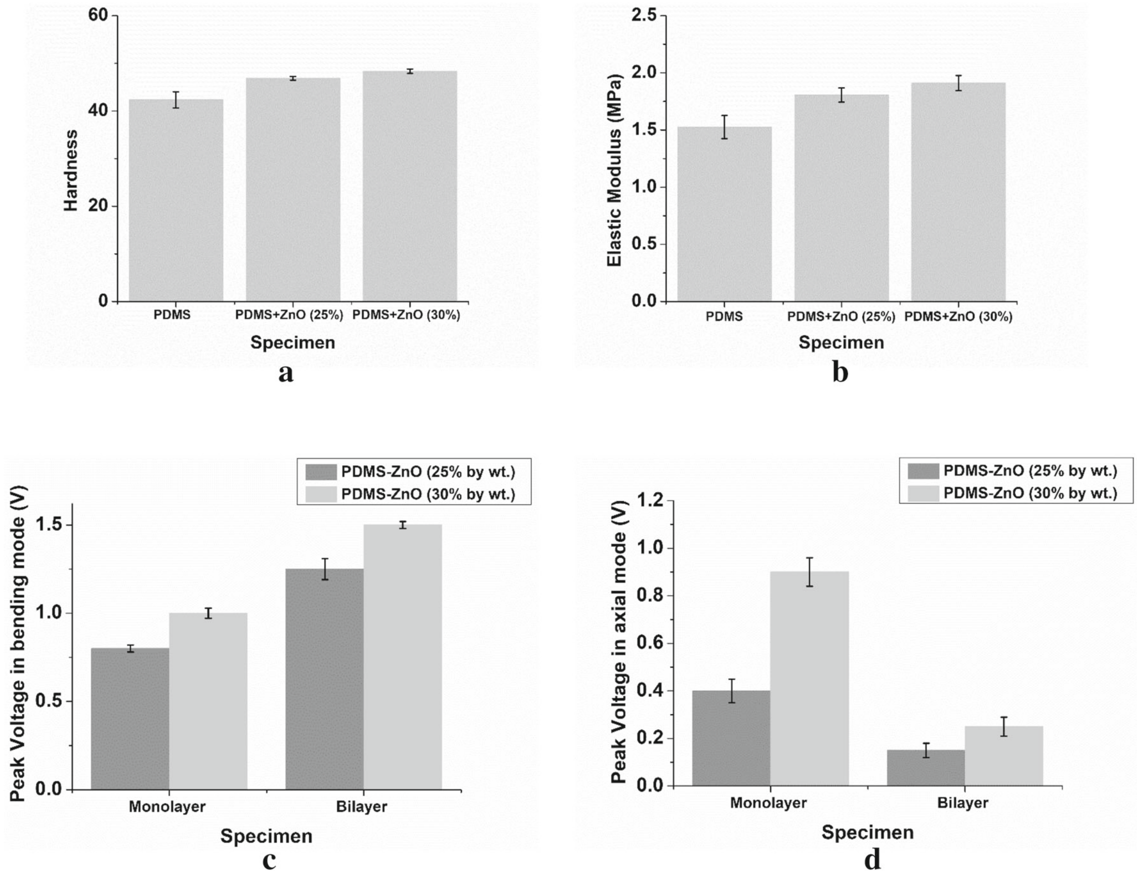

Figure 3. (**a**) Hardness of pure PDMS and PDMS–ZnO composites measured using Shore-A hardness tester. (**b**) Elastic modulus of pure PDMS and PDMS–ZnO composites calculated from the hardness values. (**c**) Peak voltage generated from the piezoelectric composite specimen in the bending mode of operation. (**d**) Peak voltage generated from the piezoelectric composite specimen in the axial mode of operation.

bilayer composite structure should produce less output voltage than that of monolayer structure. However, we found that the peak voltage generated from the bilayer composites to be higher than that of monolayer composites in the bending mode (figure 3c). For instance, peak voltage from 30 wt% ZnO monolayer and bilayer composite specimens are 1 and 1.5 V, respectively. One possible explanation for this could be that the bilayer composite is more flexible than the monolayer composite and hence produces higher output voltage for the same value of applied stress. However, we did not perform any additional tests to verify it. Furthermore, the peak voltage from the composites with 30 wt% ZnO is slightly higher than those of 25 wt% ZnO, independent of the configuration of the specimen. However, in axial mode, peak voltage from the monolayer composite specimen is about 3 times higher than that from bilayer composite specimen with the same ZnO content (figure 3d). This is a direct effect of the presence of insulating thin PDMS substrate along the direction of applied force for bilayer composites.

Previously, a piezoelectric composite made of PDMS and herbal zinc oxide has been reported in the literature [21]. A comparison of this composite with the PDMS + ZnO composite of monolayer configuration reported in this study is given in table 2. The piezoelectric strain coefficient d_{33} for the monolayer composite in this study is approximately calculated based on the axial mode testing. The d_{33} value for the PDMS + ZnO composite developed in this study is only slightly lower than that of PDMS + herbal ZnO oxide composite prepared using a relatively long and complicated fabrication process (table 2). Furthermore, the composite developed in this study is almost 10 times more flexible than the previously reported composite in the literature. This flexibility has implications in the use of piezoelectric composite in wearable sensors.

The elastic moduli of five different muscles corresponding to a relaxed and tightened fist conditions are shown in figure 4a. It can be observed that the elastic modulus of each of the muscles increases by 1.5 to 2 times after the tightening of the fist. This is expected as each of these muscles stiffens after the tightening of the fist. It is important to note that the change in the elastic modulus of muscles is of the order of few hundreds of kPa.

Figure 4b and c shows the change in the output voltage of piezoelectric composite for an increase in elastic modulus of biceps brachii and flexor carpi muscles, respectively. The

Table 2. Literature comparison of the developed PDMS + ZnO monolayer composite.

Parameter	PDMS + ZnO composite (this study)	PDMS + herbal ZnO composite (literature)
ZnO content	30 wt%	30 wt%
Fabrication technique	Addition of commercial ZnO microcrystalline powder to PDMS base and curing agent followed by curing	Mixing of synthesized herbal ZnO powder to PDMS base and curing agent followed by half-curing
Young's modulus	∼1.8 MPa	∼16 MPa
Device structure	Al–(PDMS+ZnO)–Al	PDMS–Cu–(PDMS+ZnO)–Cu–PDMS
Piezoelectric layer thickness	2 mm	0.5 mm
d_{33}	12.35 pm V^{-1}	29.76 pm V^{-1}
Application	As a sensor in muscle stiffness measurement	In energy harvesting applications

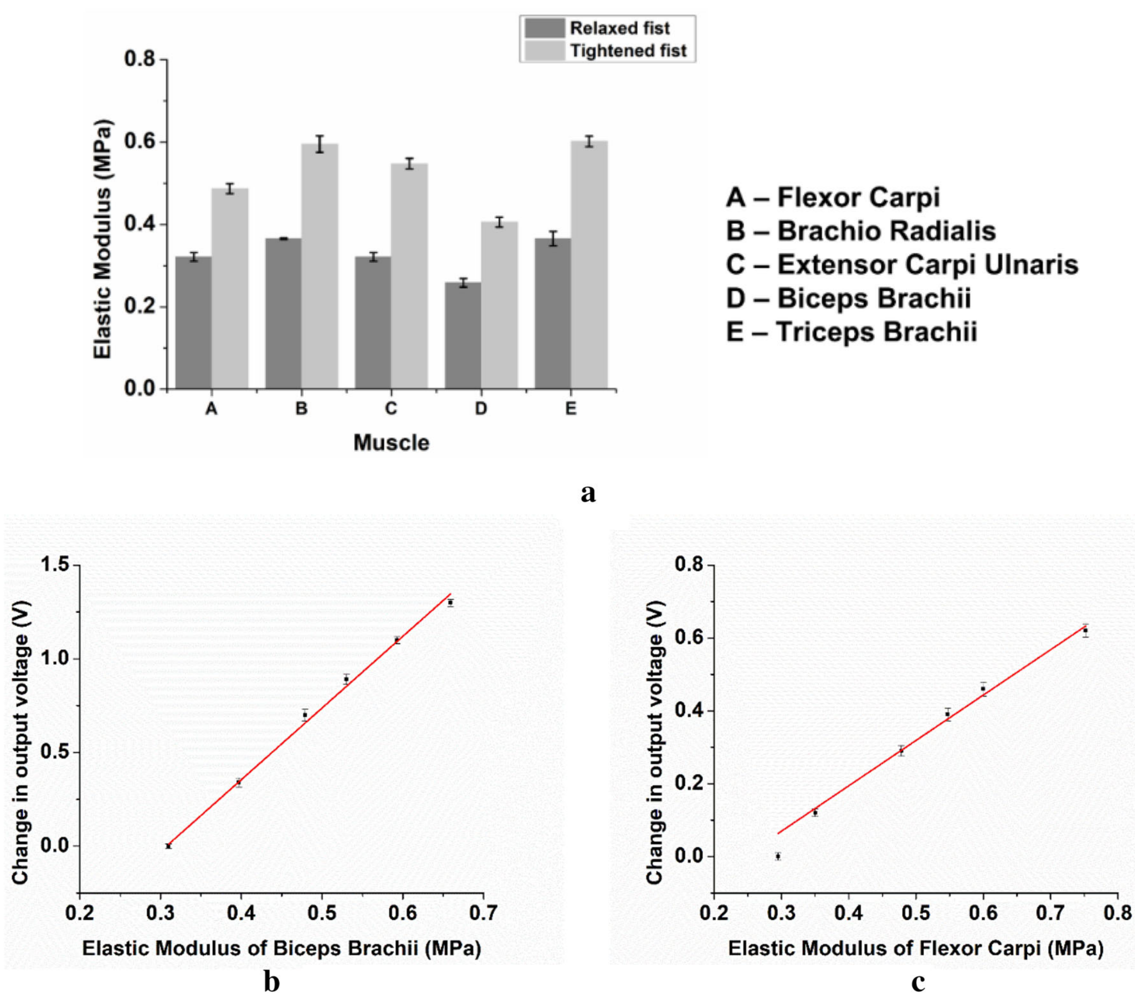

Figure 4. (**a**) Change in elastic modulus of different muscles with the tightening and relaxing of the fist. (**b**) Change in the output voltage of sensor *vs.* elastic modulus of biceps brachii muscle. (**c**) Change in the output voltage of sensor *vs.* elastic modulus of flexor carpi muscle.

experimental data followed a linear variation in both the cases, except for one value for the flexor carpi muscle. The change in elastic modulus due to the applied load in the range of 0–10 kg is almost equal for both the muscles and is in the order of few hundreds of kPa. The sensitivity of muscle activity measurement is defined as the slope of the fitted line. The sensitivity of muscle activity measurement for biceps brachii (3.826 V MPa^{-1}) is about three times of that for flexor carpi (1.245 V MPa^{-1}). An explanation for this can be given by comparing the change in the cross-section area of both the muscles for the same loading. For the same load, the increase in cross-section area due to contraction of biceps brachii is higher than that of flexor carpi. So, for the same lifted weight, strain in the piezoelectric composite is larger in the case of activation of biceps brachii. Hence, the change in output voltage from the composite is also larger during the activity of the biceps brachii muscle. For the applied load range, the behaviour of the piezoelectric composite is fairly linear and non-hysteretic. However for a higher range of elastic modulus (stiffness) of muscle, non-linearities and hysteresis would have a significant contribution to the voltage response of the piezoelectric composite.

4. Conclusions

In summary, flexible piezoelectric composites of PDMS (matrix) and ZnO (filler) are developed and tested for their mechanical and piezoelectric properties. The hardness and elastic modulus of the composites did not increase significantly ensuring that the softness and flexibility of the sensor are maintained. The piezoelectric output in bilayer composites is found to be higher than in monolayer composites in the bending mode. However, the piezoelectric output in axial mode is found to be higher for bilayer composites. For both the modes, the peak output voltage is higher from composites with 30 wt% ZnO than those with 25 wt% ZnO irrespective of the configuration.

The change in the output voltage of the sensor is found to be varying linearly with the elastic modulus of biceps brachii and flexor carpi muscles, corresponding to the applied load in the range of 0 to 10 kg. The sensitivity of muscle activity measurement for biceps brachii is found to be 3.826 V MPa^{-1} and is higher than the sensitivity of 1.245 V MPa^{-1} for flexor carpi due to a larger change in cross-section area of biceps brachii than flexor carpi muscle for the same applied load. A poling method for this piezoelectric composite material needs to be developed, so as to improve its response voltage to the external stimuli. Also, studying the viscoelastic behaviour of the composite and correction for nonlinearities and hysteresis in the piezoelectric response need to be addressed in future study.

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