# Biocompatible Gel-free Coconut-Oil and Carbon Black electrodes for ECG and Respiration measurements

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***Abstract*— The current state of the art in telemedicine has increased the interest in long term monitoring of physiological and bioelectric signals. This motivated the development of materials and techniques for the fabrication of biocompatible, user and environmentally friendly alternatives to conventional resistive wet electrodes. Here we report a method for the fabrication of dry flexible and stretchable electrodes based on Coconut-Oil and Carbon Black for the monitoring of electrophysiological signals without conductive gels. The highly stretchable material shows a specific resistance $\rho$ down to $33.2 \pm 12.3\,\Omega\,\mathrm{m}$, high conformability, and a stretchability up to $1500\,\%$. The epidermal electrodes were used to record Electrocardiographic (ECG) signals and measure respiration in a 3-lead configuration and compared to commercial wet electrodes. Even after being elongated by $100\,\%$ for 100 stretch/release cycles, a reliable recording of the QRS-complex is demonstrated without the need for any contact enhancer or substances that cause skin reaction, demonstrating the potential use of this material for long term ECG monitoring applications.**

***Index Terms*— Biocompatible electrodes, Dry electrodes, ECG electrodes, Flexible electronics,health monitoring**

## I. Introduction

THE development of novel fabrication techniques and advanced materials has fostered the creation of flexible electronic systems [1]. This has promoted the establishment of innovative applications such as wearable sensor arrays [2], smart textiles [3], or artificial skins [4] to monitor e.g. motion [5], shape [6], or health [7], while being unobtrusive, comfortable, and able to conform to the human body [8]. Such systems have to be flexible, elastic, and biocompatible to properly adapt to the skin's movement and to provide a long lasting interface [9]. These properties are of great importance, specially for electrical measurements which rely on electrodes that are in close contact with the skin [10] to acquire e.g. electrophysiological signals. Electroencephalography (EEG) [11] and Electrocardiography (ECG) are the most well known examples. These signals can be used to diagnose heart diseases, monitor health [12] and physical activity, or for human identification [13], [14]. Biopotentials are traditionally acquired with adhesive wet silver-silver chloride (Ag/AgCl) electrodes, but these can not be used continuously for more than 48 hours [15]. This is because the traditional contact enhancing conductive gel tends to dry, causing contact problems and skin irritation [16]. To prevent the use of material that dries after a specific period, metal dry electrodes have been developed that do not require electrolyte on the skin-electrode interface. However, the skin interface present potential variations in the millivolts range as a result of skin movements [17]. This has encouraged the development of new techniques and alternative materials for the fabrication of dry electrodes; some examples are the networks of flexible resistive ECG with silver nanowires [18], Carbon nanotubes or AgNPs in a PDMS matrix [19], [20], organohydrogels synthesised with ammonium persulfate, acrylamide, and acrylic acid [21], or hydrophilic textiles, clad with reduced Graphene oxide (rGO), obtained by mixing graphite powder with sulfuric acid, potassium permanganate [22]. The fabrication of these electrodes requires the use of low viscosity

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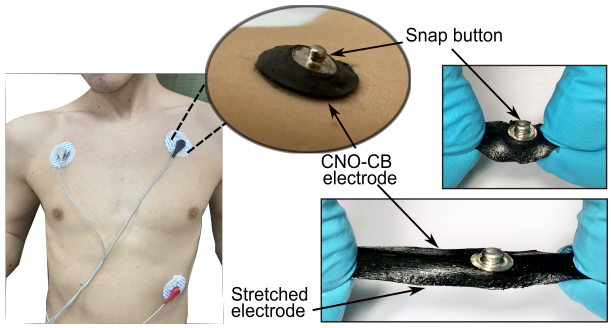


Fig. 1. Ecofriendly and biocompatible stretchable ECG electrodes fabricated with a Coconut-oil-Carbon-Black-silicone elastomer.

AgNW ink with a high voltage (1.5 kV) electrohydrodynamic printing [18], or mixing organic solvents by ultrasonication for the synthesis of MWCNT [19]. The fabrication of hydrogels needs special processes such as the stirring of chemicals for over 24 h with nitrogen protection and iced water bath sonication [21]. The rGO textiles require graphite powder, sulphuric acid, and potassium permanganate mixed in ice bath and multiple centrifugation phases [22]. The use of these materials and equipment increases the complexity and cost of the fabrication processes. Nevertheless, the resulting electrodes either showed reduced peak amplitude compared to gelled electrodes or required the use of a wet reference electrode [18]. Therefore, it is necessary to find alternative materials which are biocompatible and biodegradable and that can be processed with simple and low-cost methods for the fabrication of easy to use biocompatible electrodes with low contact resistance that do not dry or irritate the skin. Ideally, the electrodes should be used in long term monitoring applications, where they are required to withstand strains induced by deforming skin ($\approx$70 %) [23]. Such deformation generates changes in the skin-electrode interface as a result of the variations in the contact area; these variations are the main cause of motion artifacts on the potential across the epidermis on the electrode-skin interface model that have more influence in dry than in gelled electrodes [24].

In this article we propose the use of coconut-oil (CNO) based elastomer composites with a high concentration of Carbon Black (CB) particles to fabricate highly stretchable dry electrodes to perform ECG measurements and respiration detection (Figure 1). As reported in previous works, a soft and highly stretchable biocompatible material with low specific resistance can be fabricated by mixing CNO with CB in an elastomeric matrix [25], [26]. Mixing CB and Ecoflex only by stirring will generate a very low conductive material, therefore a dispersant is required. To address this, the first stage of the process consists of mixing CNO-CB to create a solution, where the coconut oil has been used as a carrier fluid to disperse the CB particles, replacing volatile toxic solvents. The proposed fabrication is a straightforward method, does not involve complex or expensive equipment, reducing the fabrication cost, promoting the production of conformable dry electrodes for the acquisition of biopotential signals. This will enable the use of this technology in areas with limited resources. Additionally, no toxic or otherwise harmful materials are used. The suitability of the created composite for recording biopotentials is demonstrated by monitoring the heart rate and respiration detection. The electrical and mechanical properties of the material were characterised and its suitability for ECG measurements was compared with state of the art Ag/AgCl dry and gelled electrodes using a commercial ADS1292R ECG evaluation kit.

## II. Experimental Procedure

### A. Electrodes Fabrication.

Figure 3 shows the fabrication process of the electrodes. The composite material was prepared using silicone elastomer Ecoflex (00-30 Smooth-on, Pennsylvania, United States), Coconut-Oil (CNO) (Pipkin, London, UK), and Carbon Black (CB) (Vulcan P, Cabot, Boston, Massachusetts, United States). Theses components were mixed with four different ratios, CB 7.5%= 10 : 5.5 : 1.3, CB 10.5%= 10 : 5 : 1.7, CB 11.3%= 10 : 4.8 : 1.9, and CB 13.2%= 10 : 4.5 : 2.2. Firstly, the CNO was melt at 37 °C. The CB was added afterwards and stirred for 0.5 h at a constant temperature (<37 °C) in a hot bath to prevent the material from lumping. After this time, part B of Ecoflex was added and stirred for 5 h. Ecoflex part A was incorporated afterwards and stirred for a further 0.25 h. After the mixing, the solution was degassed in a vacuum chamber. The solution was then poured in 10 mm radius and 1 mm depth circular 3D printed moulds. The electrodes are removed from the mould after 24 h and snapped into a snap button; the snapping pressed the CNO-CB material, forming a suction cup. This improves the electrodes' attachment to the skin by negative fluid pressure with partial vacuum. The electrodes have a contact area of $\sim$300 mm$^2$.

### B. Data acquisition circuit.

A ProtoCentral ADS1292R ECG/Respiration breakout board was used to interface the electrodes. The board was connected to an Arduino board to get the ECG signals. Figure 2 shows a schematic of the data recording system. The ADS1292R board has a differential programmable gain amplifier (PGA), as well as an ADC and voltage reference, with a 0.5 GΩ input impedance. For this analysis, the board was configured to use the internal 2.42 V reference and a sampling rate of 125 Hz with a gain of 6 V/V. From the acquisition, the first 8 bits of the raw 24-bit ADC data were discarded. As the manual suggests, a 161th order 40 Hz low pass filter was used in software combined with the IC's internal oversampling above the set data rate and a digital down sampling to reduce the output rate of the data. To reduce the common mode noise in the ECG acquisition, a Right Leg Drive (RLD) circuit was used. This circuit drives the body with the inverted common mode signal, attempting to actively cancel the common-mode interference. This signal is generated internally by the IC. As this feedback introduces additional noise, a 3 MHz RC filter was used in the IC.

ECG measurements were taken and compared with gelled and dry electrodes (ACCRINGTRD001-5-10, biosignalsplux, S.A. Lisbon, Portugal [27]), and the CNO-CB with the three different concentrations using Einthoven's 3-lead configuration [28]; this configuration is generally used to record the

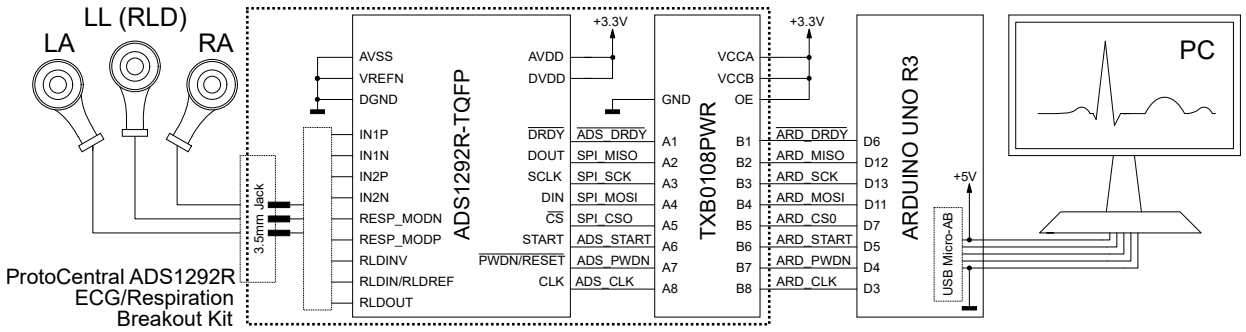


Fig. 2. ECG/respiration data recording system. The 3 leads are plugged in the ADS1292R ECG/respiration breakout board. An Arduino UNO board is used to send the digitised data from the ADS1292R to the PC over a USB COM port.

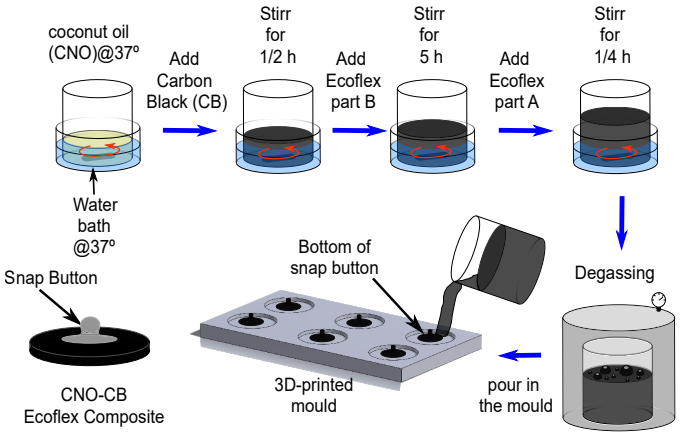


Fig. 3. ECG electrode fabrication process. Coconut Oil is heated at $37\,^{\circ}\mathrm{C}$ and mixed with carbon black. Ecoflex is added afterwards and stirrer. The solution is then degassed an poured in the moulds to set.

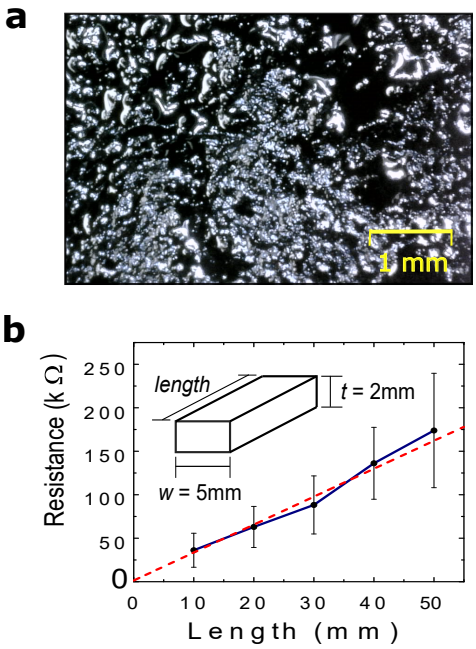


Fig. 4. Micrograph and average resistance. a) Surface micrograph of the conductive Coconut-oil composite electrode. b) Measured average resistance with standard deviation (error bars) and linear fit (red dotted line).

potential between both arms and between the left leg and each arm, allowing the measurement of a differential reading with respect to a neutral point, which is the left leg. To prove the feasibility of the electrodes for ECG monitoring, the Einthoven's 3-lead configuration was used on the chest. The electrodes were placed equidistantly to improve the results, as illustrated in Fig. 7b inset. The skin was only cleaned with Isopropanol alcohol and self-adhesive pads were used to place electrodes in the specified position; No further preparation on the skin nor contact enhancing substances were used.

Respiration detection was done with impedance pneumography. Impedance pneumography is a technique used to measure breathing rate and volume through a small change of impedance across the chest cavity [29]. Respiration monitoring was performed using Einthoven's 3-lead on chest configuration. The measurements were performed with the ADS1292R ECG/respiration board using the same setup as the ECG recording above explained.

### *C. Electrodes characterisation.*

The resistivity and impedance of the electrodes were analysed to determine their feasibility for the acquisition of ECG signals and respiration detection. The specific resistance and the contact resistance were analysed using three sets of samples with the same width ($5\,\mathrm{mm}$) and thickness ($2\,\mathrm{mm}$) but 5 different lengths (10, 20, 30, 40 and $50\,\mathrm{mm}$).

Five glass slides were prepared with two copper tapes attached to its surface and separated (10, 20, 30, 40 and $50\,\mathrm{mm}$). Afterwards, each sample was placed on the glass slide with the bottom surface contacting the copper tapes. The resistance was then measured between the copper tapes with a digital multimeter (34465A, Keysight, Santa Rosa, CA, USA). The stability of the electrodes' resistance under tensile strain was obtained measuring the resistance while each electrode was subjected to 100 stretch cycles over a range from 0 to $100\,\%$, which is more than the maximum stretchability of the skin, at a strain rate of $171\,\mathrm{mm/min}$. After the tests, ECG and respiration measurements were carried out again to compare pristine and stretched electrodes. Similarly, the maximum tensile strain an electrode can sustain was analysed with a maximum stretchability test while measuring the resistance with the multimeter. Both tests were performed in an automated stretch system made of a linear motion stage, a stepper motor, and a motor driver that were controlled and interfaced with an Arduino UNO board.

The characterisation of the skin-electrode interface is of interest to determine the correct electrical information to obtain a clear ECG signal. ECG profile distortion can occur as a result of a phase shift introduced to the acquired signal by the skin-electrode interface [30]. A high electrode-skin interface impedance increases the thermal noise, which in turn reduces the usable resolution of the signal. Therefore, this interface was characterised by measuring the impedance. An equivalent circuit model of the skin-electrode is shown in figure 5. This model includes the capacitance across the electrode-CNO interface $C_1$, Charge transfer resistance $R_1$, the capacitance induced by the stratum corneum layer $C_3$, the capacitance induced by the sweat glands and ducts $C_4$. $R_3$ and $R_4$ accounts for the resistance of the soft tissue layer, while $R_5$ is the overall resistance of the tissue underneath the epidermis layer. The model also includes the electrode potential difference, $E_1$ of the CNO-CB solution, while $E_2$ and $E_3$ are the potential differences resulting of the differences in ion concentration of the subcutaneous tissue [31]. Based on this equivalent circuit, the skin-electrode impedance of the CNO-CB electrode $Z_{CNO\text{-}CB}$, can be calculated with equation 1.

$$Z_{CNO-CB} = R_2 + \frac{R_1}{1 + j\omega R_1 C_1} + R_5 + \frac{R_3}{1 + j\omega R_3 C_3} + \frac{R_4}{1 + j\omega R_4 C_4} \quad (1)$$

Electrical characteristics were obtained by measuring the voltage drop when a 1V sine wave was applied to a voltage divider with two electrodes and a $1\text{k}\Omega$ resistance. The electrodes were placed $5\,\text{cm}$ from each other on the forearm with no skin pre-treatment. The applied frequency was varied from 10Hz to 10MHz using a 2-wire impedance analyser board ($50\,\text{pF}$ to $500\,\mu\text{F}$ capacitance range, $10\,\mu\text{Hz}$ to $1000\,\text{mHz}$ inductance measurement range, and $1\,\text{Hz}$ to $15\,\text{MHz}$ excitation frequency range, Diligent Inc., Pullman, WA), plugged to a Discovery 2 digital oscilloscope and logic analyser (Diligent Inc., Pullman, WA). The impedance analyser board was configured to use a $1\,\text{k}\Omega$ resistance. 1000 samples were taken at each frequency and the measurements were taken sequentially from low frequency to high frequency.

## III. Results and Discussion.

Figure 4a shows an optical micrograph of the material's surface. The resulting material is a non-homogeneous oily CNO-CB suspension embedded in a porous elastomeric conductive matrix. This combination created a rough oily surface with CNO-CB oil droplets smaller than $1\,\text{mm}$, which reduced the contact resistance. Figure 4b shows a plot of the average resistance of the $10.5\,\%$ CNO-CB polymer at five different lengths. The resistance was measured from the copper tapes in contact with the end of the samples. There is a linear relationship between resistance and length with the standard deviation increasing for longer samples. These measurements are not affected by the contact interface, as expected, demonstrating a negligible contact resistance, a desirable feature in ECG electrodes. This indicates a non homogeneous dispersion of the conductive filler on the polymeric matrix. The resulting intrinsic resistivity is $\rho = 33.2 \pm 12.3\,\Omega\,\text{m}$ for $10.5\,\%$wt concentration, while the $7.5\,\%$wt concentration resulted in a significantly higher $\rho$ of $145.8 \pm 71.2\,\Omega\,\text{m}$.

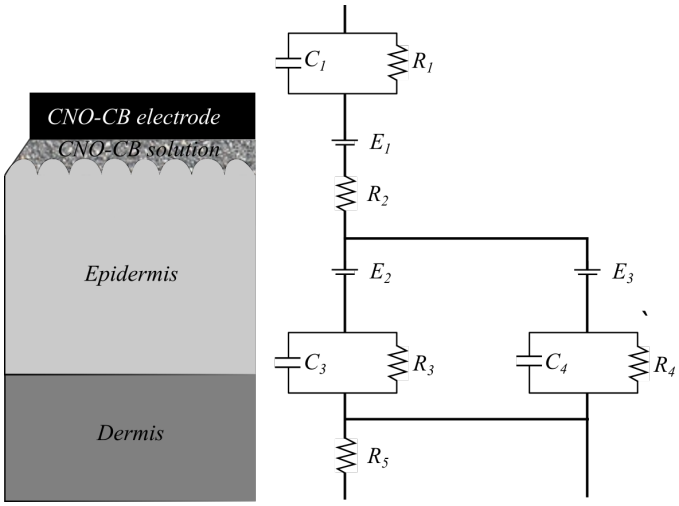


Fig. 5. Skin-electrode interface model and equivalent electric circuit.

### A. Respiration measurements.

The suitability of the electrode for recording biosignals was analysed by taking respiration and ECG measurements. The results of the respiration measurements are displayed in Figure 6. The results show the phase difference between the wave sent through the chest and the original signal. The data was unwrapped from -$\pi$ to $\pi$ before being filtered for clarity. The raw signal from the sensor is clipped to -$\pi$ to $\pi$. This causes frequent large discontinuities and makes it difficult to visualise the signal. For clarity the signal is unwrapped by adding multiples of $\pm 2\pi$. A Band-Pass filter with corner frequencies far from the signal of interest was implemented to remove the DC offset from the unwrapped signal caused by phase drift. The absolute phase difference depends on many other factors and the breathing rate can be extracted from the frequency of the wave. Figure 6 depicts how the breath signal is larger with coconut-oil electrodes, and the further increase of CB concentration enhances the signal detection. However, this enhancement also shows an increase in the size of the artefacts. Although the results show the feasibility of the electrodes to measure respiration, the higher resistance and sensitivity of the electrodes developed may require different circuit design/conditioning. The increased sensitivity of the carbon based electrodes may have been caused by the higher resistance. For a similar capacitance of the chest, a higher resistance will cause a larger phase difference in the signal.

### B. ECG measurements.

The ECG recordings taken with the CNO-CB electrodes can be observed in Figure 7, which displays ECG signals taken with the three different concentrations of CNO-CB, being compared with the Ag/AgCl dry and gelled electrodes. In this figure it can be clearly seen that the electrodes with $11.3\,\%$wt CB provide a better signal quality (figure 7c)

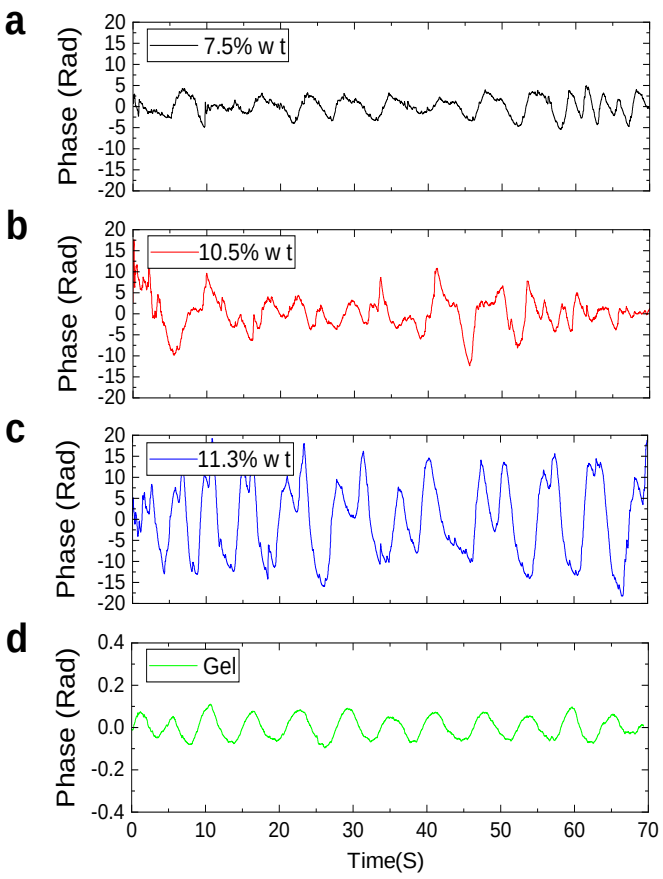


Fig. 6. Comparison of respiration signals taken in a Chest 3-lead configuration with four different materials) 7.5 %wt CB concentration, b) 10.5 %wt CB concentration, c) 11.3 %wt CB concentration and d) Ag/AgCl reference wet gel electrodes.

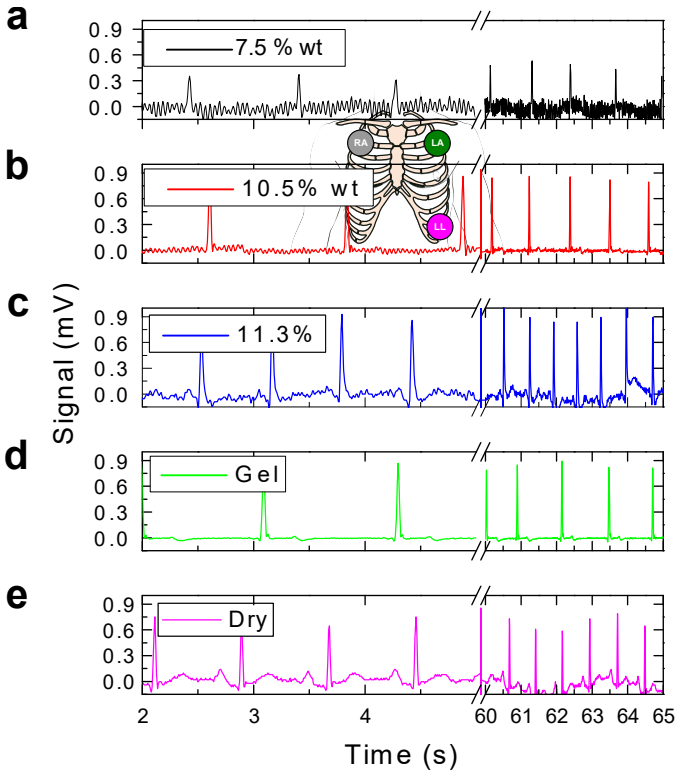


Fig. 7. Comparison of ECG signals taken using a chest 3-lead configuration with three different materials, a) 7.5 %wt CB, b) 10.5 %wt CB, c) 11.3 %wt CB, d) Ag/AgCl reference wet gel electrodes, and e) Ag/AgCl dry electrodes.

than with the other two concentrations of CB (10.5 %wt and 7.5 %wt). This clearly demonstrates a proportional effect of the CB particles on the signal acquisition, which is the effect of decreasing the contact resistance when increasing the CB particles concentration. The 11.3 %wt CB electrodes exhibit an amplitude of ≈0.98 mV and those with 10.5 %wt CB concentration exhibit a slightly lower signal amplitude of ≈0.85 mV. In both cases, the QRS-complex are similar to those of the wet electrodes, but is more clearly defined and less noisy in the electrodes with a higher CB concentration than the electrodes with lower CB concentration. This shows a clear difference in the signal amplitude and noise. The comparison of the QRS complex demonstrates that although the signal acquired with the 11.3 %wt CB electrodes has a slightly higher amplitude (0.99 mV) than that acquired with the gelled electrodes (0.95 mV), the latter is more neat because of the improved contact resistance. This can be appreciated by analysing the Signal Amplitude to Noise Amplitude Ratio. The electrodes with 11.3 %wt CB exhibited a Signal Amplitude to Noise Amplitude Ratio of 49.75 while that of the sample with 10.5 % wt CB was of 27.43 and that of the 7.5 %wt CB was 8.72. The latter is comparable to that of the dry electrodes which is 7.76. In comparison, the ratio of the gel electrodes is 62.65. Although the amplitude of the signal of the 11.3 % wt CB electrodes is similar to that of the gel electrodes which have a better ratio and less noisy signal, as it can be observed in figure 11. It is worth noting that the measurements were taken with the IC designed for the gel electrodes and no filter was applied while taking the CB electrodes signal. Also, no contact enhancers (e. g. conductive gel) were used during the tests.

### C. Electrical characterisation.

The electrical behaviour of the material was additionally evaluated by comparing the skin contact impedance. However, the electrical characteristic can be different for each person and circumstance.

It can be observed in figure 8 that although the electrochemical model for the gel electrodes is not a suitable for the CB electrodes, they still measuring the respiration rate effectively. At low frequencies (10 Hz) the impedance of the 11.3% Carbon electrode is $(408 - 494j)$ kΩ compared to the gel electrode with $(1299 - 1258j)$ kΩ.

### D. Mechanical characterisation.

The mechanical behaviour of the material was evaluated by performing tensile strain test, analysing the maximum tensile strain the electrodes can sustain and their long term resistance to mechanical deformation. Figures 9a and 9b show the results of representative samples with 10.5 %wt and 11.3 %wt CB respectively, under a cyclic strain that ranges

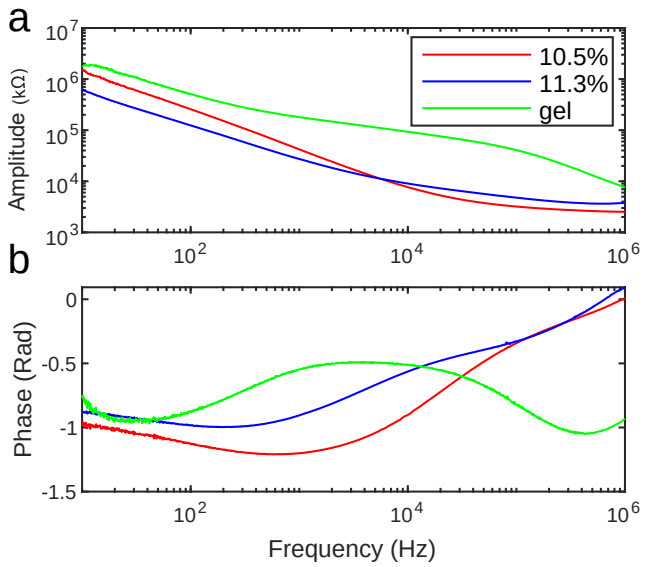


Fig. 8. Impedance for CNO-CB with $10.5\,\%$, $11.3\,\%$ CB and Ag/AgCl gel electrodes. Each pair was separated $5\,\mathrm{cm}$ on skin.

from 0 to $100\,\%$ at a strain rate of $171\,\mathrm{mm/min}$. Figure 9a illustrates how the resistance of the electrode in relaxed state is hardly influenced by the cyclic strain. It can be observed that

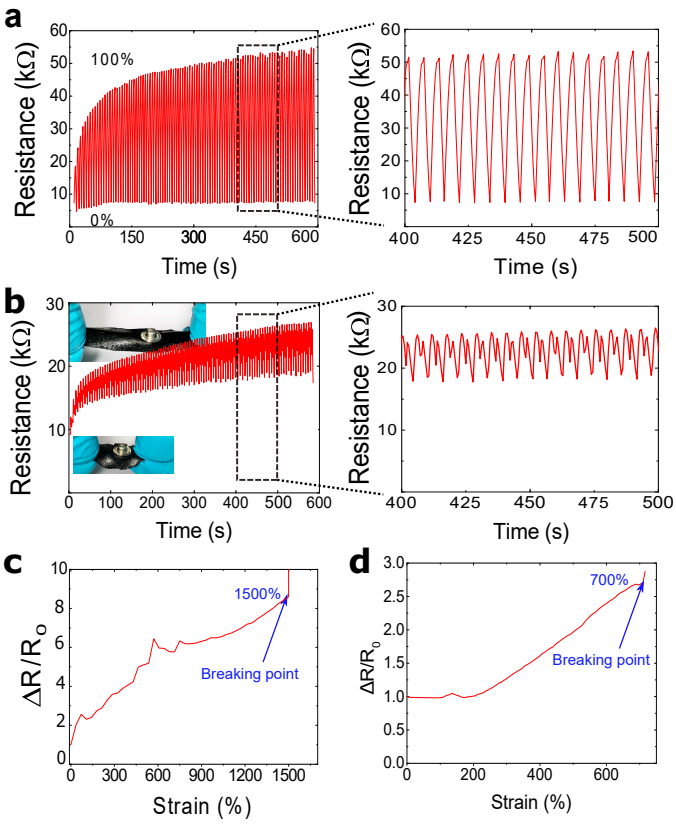


Fig. 9. Tensile strain test on the CNO-CB electrodes. Cyclic strain test under 100 stretch/release cyclic test at a strain rate of $171\,\mathrm{mm/min}$ on electrodes with a) $10.5\,\%$wt and b) $11.3\,\%$wt CB concentration. Tensile strain test to rupture on electrode with a c) $10.5\,\%$wt and d) $11.3\,\%$wt CB concentration.

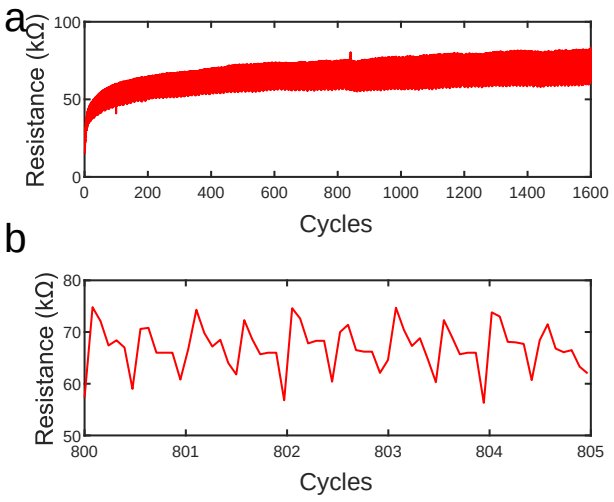


Fig. 10. 1600 cycles Strain test at $100\,\%$ of the CNO-CB with a $11.3\,\%$ CB concentration.

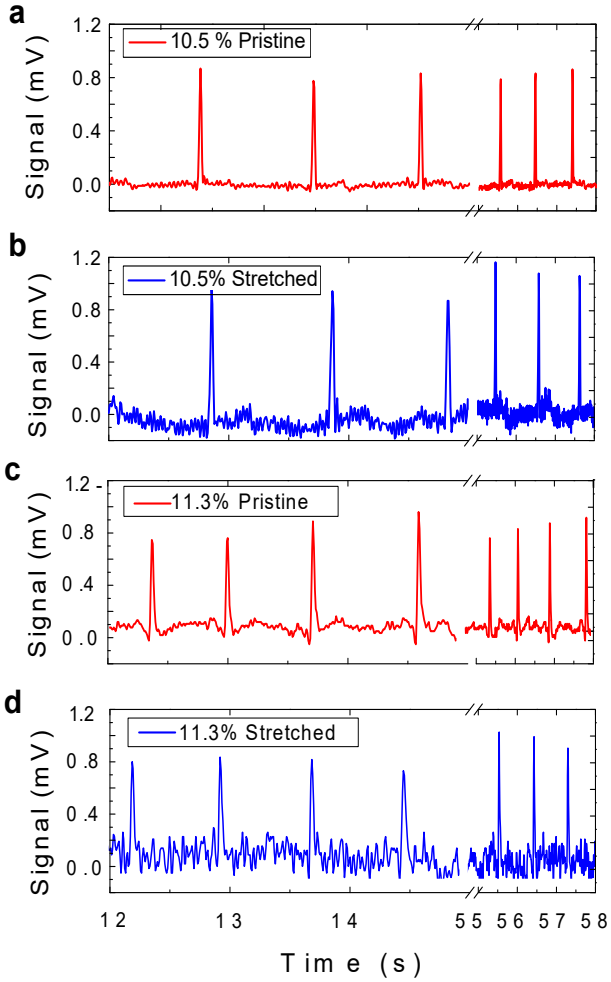


Fig. 11. ECG signals using the CNO-CB electrode. ECG signals acquired with the a) pristine electrode and b) with the electrodes after cyclic test.

the resistance at $0\,\%$, $\mathrm{R_0}$, was $5.85\,\mathrm{k\Omega}$, representing $0.015\,\%$ change, with an average $\mathrm{R_0} = 7.26 \pm 0.66\,\mathrm{k\Omega}$. On the other

hand, the resistance at 100 %, $R_{100}$, was 54.5 kΩ, three times its initial value (18.4 kΩ). The sample with 11.3 %wt CB shows a higher changes in the initial and final resistances after 100 cycles. Its resistance at 0 %, $R_0$, was 10.4 kΩ, and increased to $R_{100}$=21.4 kΩ. Similarly, its resistance at 100 %, $R_0$, was 11.9 kΩ, and increased up to $R_{100}$=25.8 kΩ. Despite the increase in the resistance, the electrode with 11.3 %wt CB exhibited higher stability during the cyclic tests (Figure 9b), observing an average of 4.5 kΩ change in resistance. In Figure 9b a double peak is observed at 100 % strain. First, there is an overshoot when reaching the maximum strain (100 %) followed by a brief relaxation while at this state. Then, another increase in the resistance is observed when the strain starts to going back from 100 % to 0 %. In Figure 9a a similar behaviour is observed but is less clear because of the range of the resistance presented. In both cases, the behaviour is the result of the reorientation of both conductive particles and the molecules of the elastomer caused by stress relaxation [32]. The maximum strain supported by the developed material was determined by stretching a sample until the breaking point while recording the resistance. Figures 9c and 9d show the increase in resistance with strain for the samples with 10.5 %wt CB and 11.3 %wt CB respectively. The former yielded an ultimate strain of 1500 % with a change in resistance of 8, while for the latter was 700 % with a change in resistance of 2.7. The initial value of the resistance of the sample with 11.3 % was of 68.2 kΩ increasing to 70.4 kΩ at 200 % strain, denoting an increase on the resistance of 21 %. A longer cyclic test was performed on a different sample with 11.3 % CB concentration, simulating real time monitoring for more than 24 h [20]; Figure 10a shows the tests and it can observed that the trend continues and the resistance increases further. After 1600 cycles, the sample showed a difference in the resistance of 40.2 % (23.6 kΩ) between stretched (82.2 kΩ) and unstretched (58.6 kΩ) states. Comparing both samples after 100 cycles, it was observed a difference of 60.1 kΩ, being higher in the second sample.

Finally, further ECG measurements were performed to demonstrate the usability of the electrodes after the 100 cycles stretching test. The results are displayed in Figure 11. Although a slight increase in the signal's amplitude was observed after the cyclic test (Figure 11b), the QRS-complex are still clear and well defined, denoting a small effect of stretching on the soft coconut-oil electrodes performance.

## IV. Conclusions.

Bio-compatible stretchable dry ECG electrodes were fabricated using coconut-oil and carbon black. The material can withstand large strain values as well as repetitive stretching, remaining effective even after being elongated by 100 % for 100 cycles. The material has electrical properties and a structure comparable to that of tunnable piezo-resistive polymer foams with limited elasticity (<60%) [20], [33], [34]. The electrodes have demonstrated to be suitable to measure ECG signals and impedance pneumography without the need for contact enhancer or conductive gels. However, they showed a slightly higher impedance and artefacts compared with the conventional Ag/AgCl electrodes. Dispersing CB particles in the coconut oil generates an oily suspension. Therefore, we can infer that if the electrodes remain in the same place during long term measurements, high changes in the resistivity of the samples as a result of drying might be considered unlikely. This can be of advantage for long term monitoring of biopotentials. Future work will include the signal integrity improvement and the analysis of the electrodes' performance and stability in long-term measurements. The main advantages of the CNO-CB electrodes is that they are environmentally friendly and produced with a simple and inexpensive fabrication technique. Although the material leaves oily residues on the skin, these can be removed easily by scrubbing and soapy water. Despite of this, the electrodes have the potential to be used in long-term monitoring applications, as they could work reliably without the need of additional electrolyte or gels, even after the cyclic test. Coconut oil has a high content of essential vitamins and fatty acids and is widely used as skin moisturiser [35], therefore it could help to prevent problems such as skin irritation during ECG measurements.

## References


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