Supplementary Materials for

Self-adaptive cardiac optogenetics device based on negative stretching-resistive strain sensor

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The phenomenon of negative relative resistance variation of the composites with dispersed fillers is attributable to a combination of the influences of the conductive fillers and the polymer matrix. The NL used in this work facilitates the formation of multiple conductive paths, thus exhibiting a specific electrical response to strain. Here, we propose an equivalent circuit connecting the material changes of the composites to their electrical characteristics. The total resistance of the CNL membrane is decomposed into three parts to describe the three types of conductive situation when the membrane is stretched. Based on the above analysis, the observation of the negative stretching-resistive effect has been proved theoretically in details.

Conductor-insulator composites, which are polymeric materials with uniform dispersed conductive fillers, are proved to show electrical properties by transferring electrons between the conducting fillers of a percolating network. For such composites, the total resistance is the result of three main electron conduction mechanisms, including (I) \( R_i \): the intrinsic resistance of the conductive filler, (II) \( R_{\text{tun}} \): the tunneling resistance of two adjacent fillers that realize electron tunneling (37) and (III) \( R_{\text{cont}} \): the direct contact resistance between two connected conductive fillers. The specific influences of the above three resistances in negative stretching-resistive composites are discussed in detail in the following sections.

I. Intrinsic Resistance of the Carbon Nanotubes \( R_{\text{cnt}} \)

In the proposed CNL membrane, the CNTs can be regarded as good conductors with a near one-dimensional structure that allows electrons to flow in the ballistic condition (38). In addition, it is well established that the Young’s modulus of MWCNTs (~0.5 GPa) (39) is about 17 times that of NL (30 ± 19 MPa) (25); so, during the tensile process, the bending and deformation of the CNTs cannot be neglected in numerical studies. The intrinsic resistance of CNT is defined as follows:

\[
R_{\text{cnt}} = \frac{1}{\sigma_{\text{cnt}}} \cdot \frac{l_{ij}}{S_{\text{cnt}}},
\]

where \( \sigma_{\text{cnt}} \) is the electrical conductivity of CNT, \( S_{\text{cnt}} \) is the cross-sectional area of the CNT and \( l_{ij} \) is the effective length between the generic points \( i \) and \( j \) of the CNT that are involved in the percolation networks. As shown in Fig. S5, when the negative stretching-resistive composite is stretched along the x-axis, the composite is forced to shrink along the direction perpendicular to the stretching due to the large Poisson’s ratio (~0.5) of the composite. In consequence, the CNTs wrapped around the NL particles move not only uniaxially but also rotationally, resulting in a reduction of \( l_{ij} \), and then, \( R_{\text{cnt}} \) decreases accordingly (40).

However, the variation of tunneling resistance \( R_{\text{tun}} \) is much higher than that of the intrinsic resistance of CNTs; so, more attention should be paid to the tunneling phenomenon in the negative stretching-resistive composite (41).

II. Tunneling Resistance of Adjacent CNTs \( R_{\text{tun}} \)

On the interpretation of the mechanism of conductive polymers, the tunneling phenomenon is a widely accepted theory, which indicates that once the distance between a pair of conductive fillers is sufficiently small, the electrons will be activated by thermal vibrations under an electric field. Consequently, the electrical charges can flow through the energy barrier caused by the tiny insulating matrix (42,43). The tunneling resistance between adjacent CNTs is estimated by Simmons’ formula (44) and is given by

\[
R_{\text{tun}} = \frac{V}{A J} = \frac{h^2 d}{4 \pi d \sqrt{2m_e \lambda}} \times \exp \left( \frac{4 \pi d}{h \sqrt{2m_e \lambda}} \right),
\]

where \( V \) is the potential difference, \( A \) is the cross-sectional area, \( J \) is the tunneling current density, \( h \) is the Plank’s constant, \( d \) is the distance between CNTs, \( e \) is the electron charge, \( m_e \) is the electron mass and \( \lambda \) is the height of the barrier. It can be inferred that \( R_{\text{tun}} \) increases exponentially as \( d \) increases. In order to emphasize the dependence of the distance and the barrier height, Eq. (S2) can be considered in a simpler form:

\[
R_{\text{tun}} = C_1 \frac{d}{\sqrt{\lambda}} \times \exp(C_2 d \sqrt{\lambda}),
\]
where
\[ C_1 = \frac{\hbar^2}{4\pi^2 e^2}, \quad (S4) \]
\[ C_2 = \frac{4\pi}{h} \sqrt{2m_e}. \quad (S5) \]

Therefore, the relative resistance change under tension is defined as follows:

\[
\frac{\Delta R_{\text{tun}}}{R_{\text{tun}0}} = \frac{R_{\text{tun}} - R_{\text{tun}0}}{R_{\text{tun}0}} = \frac{c_1 d \exp(c_2 d \sqrt{\lambda}) - c_1 d_0 \exp(c_2 d_0 \sqrt{\lambda})}{c_1 d_0 \exp(c_2 d_0 \sqrt{\lambda})}, \quad (S6)
\]

where \( d \) is the separation distance between the adjacent CNTs under the applied strain, and \( d_0 \) is the separation distance without strain (\( \varepsilon = 0\% \)). For the sake of clarity and completeness, Eq. (S6) can be reduced to the following simplified version to display the relationship of the distance variation and the tunneling resistance:

\[
\frac{\Delta R_{\text{tun}}}{R_{\text{tun}0}} = \chi \exp[c_2 \sqrt{\lambda} (d - d_0)] - 1, \quad (S7)
\]

where \( \chi \) is the ratio of \( d \) to \( d_0 \). Furthermore, the derivative of \( \Delta R_{\text{tun}}/R_0 \) is calculated, and the outcome is given as follows:

\[
\frac{\partial (\Delta R_{\text{tun}}/R_{\text{tun}0})}{\partial d} = \frac{1}{d_0} (1 + c_2 \sqrt{\lambda} d) \exp[c_2 \sqrt{\lambda} (d - d_0)]. \quad (S8)
\]

The values of \( d \) and \( d_0 \) are always positive; so, the derivative of the relative tunneling resistance change must be greater than zero, which means that the function of the relative tunneling resistance variation is a monotonically increasing function. Hence, we can infer that the necessary condition of the negative relative tunneling resistance variation is that the separation distance between adjacent CNTs under the applied strain should be smaller than that under zero strain. Considering the micorscale, it is possible for the separation distance to increase or decrease when the composite is stretched, and the total separation distance change occupying the dominant position will determine the final net resistance variation.

It is inferred that there are two main factors influencing the separation distance between conductive fillers: the concentration of CNTs and the Poisson’s ratio of the composite. On the one hand, with the increasing concentration of CNTs (also named volume fraction), the short distance between adjacent CNTs decreases substantially (41). Then, the percolation networks consisting of CNTs become denser than those with lower concentrations, providing a favorable condition for the tunneling effect. This phenomenon is enhanced in the CNL membrane because the strong bonding between CNTs and NLs causes the CNTs to concentrate in the surroundings of the NL particles rather than randomly distribute through the polymer matrix. Besides, the orientations of CNTs and NL particles lead to an arrangement along the direction of stretching, thus offsetting the result of increasing separation due to stretching. On the other hand, a numerical analysis method based on the Monte Carlo approach has been proposed to introduce the average junction gap variation (AJGV) as a quantitative description of the resistance change caused by strain (40). In particular, the AJGV is defined to exhibit the net effect of the positive and negative variations of CNT gaps and can be expressed in the form:

\[
\text{AJGV} = \frac{1}{N} \sum \Delta d_i = \frac{1}{3} \left( D + \frac{1}{2} H_C \right) (1 - 2\nu) \varepsilon, \quad (S9)
\]

where \( N \) is the number of tunneling junctions in the percolating network, \( D \) is the diameter of the CNTs, \( H_C \) is the critical tunneling shell thickness, \( \nu \) is the Poisson’s ratio of the composite and \( \varepsilon \) is the strain applied on the composite. It can be deduced that the AJGV tends to be smaller when the Poisson’s ratio of the composite is larger. Coincidentally, the NL we utilized is a kind of polymer material with a high Poisson’s ratio, causing the relative tunneling resistance variation to be negative.

**III. Direct Contact Resistance of Connected CNTs \( R_{\text{cont}} \)**

The direct contact resistance \( R_{\text{cont}} \) of the connected CNTs also contributes to the overall change of resistance. As mentioned in the section of ‘Mechanism and characteristics of negative stretching-resistive effect,’ the original conductive paths are strongly compressed by NL particles and become
more compact in the situation S1. As a one-dimensional nanomaterial, CNTs with a high aspect ratio are conducive to the formation of CNT percolation networks by overlapping each other (45). Moreover, the negative stretching-resistive composite is fabricated by a facile latex approach in which the nanomaterial is mixed with the rubber latex in a water dispersion system. Compared with other methods, this approach retains the high aspect ratio of CNTs as much as possible (46,47); so, the CNTs can overlap each other to construct more direct contact points for electron transmission.

Here, we introduce an optimized R-C parallel equivalent circuit to model the ideal negative stretching-resistive composite. Both the electrical behaviors of CNTs and NL are taken into account (41). Specifically, the total resistance of the percolation paths is associated with the intrinsic resistance of CNTs ($R_{cnt}$), the direct contact resistance of connected CNTs ($R_{cont}$) and the tunneling resistance ($R_{tun}$) (41). Here, $R_{cont}$ is ideally regarded as zero to simplify the equivalent circuit. In addition, $R_{tun}$ is parallel to the capacitance ($C_{gap}$), which models the displacement current between adjacent CNTs for the complete analysis at high frequencies (48). The resistance $R_{latex}$ indicates the dispersive behavior of the bulk polymer matrix (i.e., NL), which is much higher than the resistance of CNTs and it can be regarded as invalid path for electron transfer in the equivalent circuits. The equivalent circuits as well as the corresponding three types of conductive situation under zero strain and under tension are schematically shown in Fig. S6.

First, for S1, the equivalent circuit is shown in Fig. S6A, in which the yellow region highlighted infers to the connected path of CNTs. When the CNL membrane is stretched, the intrinsic resistance of CNT decreases (as shown in Fig. S6B) and more compact percolation networks are constructed. Therefore, it can be regarded as more parallel conductive branches of CNTs are formed. It is well established that the more parallel branches, the smaller the total resistance.

Second, with regard to S2 (as shown in Fig. S6C and Fig. S6D), the shrinkage perpendicular to the stretching direction further decreases the tunneling distance of adjacent CNTs; so, more direct conductive paths occur.

Third, Fig. S6E and Fig. S6F show the changes of S3 before and after stretching, respectively. In the original state, the CNT aggregates are separated with large gaps at Y direction; so, the conductive paths in this direction are hindered, but with the gradual increase of stretching and shrinkage, the conductive paths caused by the tunneling effect will enforce the electron transfer. Thus, it can be regarded as more tunneling resistance of CNTs appear to replace the $R_{latex}$ with extremely high resistance.

From the above analysis, it can be known that the total resistance of the CNL membrane decreases with tension because more effectively conductive paths are formed. The equivalent circuit offers an intuitive tool for the evaluation of the negative stretching-resistive strain sensor and can be utilized in the analysis of time-domain performance.
Fig. S1. Relative resistance changes of CNT weight ratio under 3 wt%. (A) Resistance change versus strain of CNT with a ratio of 1.1, 2.0, 2.5 and 2.9 wt%. (B-C) Resistance change of CNT with a ratio of 2.5 wt% and 1.25 wt% under stretching.
Fig. S2. The ultra stretchability of CNT-NL (CNL) membrane. (A) The membrane is able to be stretched up to 640%. (B) The resistance response in a strain range of 0%-640%. Photo Credit: Chunpeng Jiang, Shanghai Jiao Tong University.
Fig. S3. Positive stretching-resistive effect in multiple conductive fillers and polymers. (A-C) carbon black (CB), silver nanowires (AgNWs) and graphene as the conductive fillers doped in the same NL solution, respectively. (D-F) CNTs doped in different polymers like TPU, hydrogels and PDMS, respectively.
Fig. S4. Illustration of positive stretching-resistive effect occurs in the CNT-NL composites with low CNT concentration. (A) Sectional view of the unstretched CNT-NL membrane, which can light an LED under a certain applied voltage. (B) The stretched CNT-NL membrane prevents electrons from transferring, leading the LED to extinguish immediately.
Fig. S5. The effective length of CNTs in the CNL composites (A) before stretching and (B) corresponding changes caused by their movement and rotation after stretching.
Fig. S6. Equivalent circuits of the negative stretching-resistive film. (A-B) Changes of equivalent circuit that correspond to S1 before and after stretching. (C-D) Changes of equivalent circuit that correspond to S2 before and after stretching. (E-F) Changes of equivalent circuit that correspond to S3 before and after stretching.
Fig. S7. TEM of the islands constructed by several CNL cells and connections between islands constructed by CNT bridge.
Fig. S8. SEM of the lipid-free deproteinized natural rubber.
Fig. S9. The XRD patterns of CNL membrane before stretching and under a stretching of 30% strain, respectively.
Fig. S10. Resistance response of the CNL membrane with strain under different strain rate of 5 mm/min, 10 mm/min and 15 mm/min.
Fig. S11. Long-term test of the CNL membrane with two thousand cycles.
Fig. 12. The comparison of (a) traditional cardiac optogenetics device (36) and (b) our design including negative stretching-resistive sensors with (c) a mechanism of self-adaptive light stimulation.
Fig. S13. Schematic of the riveting structure between CNL membrane and corresponding electrodes. Photo Credit: Chunpeng Jiang, Shanghai Jiao Tong University.
Fig. S14. Luminous LEDs with isolation of Ecoflex are immersed in the water for safety check. Photo Credit: Chunpeng Jiang, Shanghai Jiao Tong University.
Fig. S15. Illustration of the injection process of ArchT virus, which employs a micro-injection pump, a 50-μL microneedle and capillary as injection source with constant velocity, together with an adjustable microneedle for myocardial injection.
Fig. S16. Photos of the *in-vivo* experiment. (A) The implantation image of optogenetics device wrapped the heart. (B) The self-adaptive circuit connected with the optogenetics device. Photo Credit: Wen Hong, Shanghai Jiao Tong University.
Fig. S17. The analog strain changes of four-channel negative stretching-resistive strain sensor array under normal heartbeats with an estimated rate of 90 times/min.
Fig. S18. The analog strain changes of four-channel negative stretching-resistive strain sensor array under heartbeats with an estimated rate of 120 times/min.
Fig. S19. The sensing voltage of a negative stretching-resistive strain sensor and corresponding digital pulse under a heart rate of 90 times/min.
Fig. S20. The ECG, intracadiac electrogram and epicardial electrogram recorded in the control in vivo experiment, in which I, II, III, avR, avL and avF are the limb leads of the electrocardiograph (ECG); CS D-2, CS 3-4, CS 5-6, CS 7-8 and CS 9-10 are the records of intracadiac electrogram; and Pen 3-4, Pen 5-6, Pen 7-8 and Pen 9-10 are waves of epicardial electrogram.
Fig. S21. Accelerated aging experiment performed in the 10× PBS solution of 57°C for three weeks. The relative resistive variation of the CNL membrane tested (A) before and (B) after the accelerated aging experiment. (C) The lighted LEDs after the accelerated aging experiment. Photo Credit: Wen Hong, Shanghai Jiao Tong University.
Fig. S22. The images of the cell proliferation and morphology of the group 1, group 2 and control group over 36 hours. HT-22 cells incubated with the CNL film sealed by Ecoflex (group 1), the bare Ecoflex film (group 2) and the control group incubated with DMEM only. All scale bars, 100 μm.
Fig. S23. The cardiac tissue is injected with AAV2/9-CAG-ArchT-GFP virus, which whitens a little bit. Photo Credit: Wen Hong, Shanghai Jiao Tong University.
Fig. S24. Zeta potential diagram of solution indicating the homogeneity of the CNL solution.
| Serial Number | Classification                                    | Ratio of CNT solution and natural latex solution | Weight ratio of CNT (%) |
|---------------|--------------------------------------------------|------------------------------------------------|------------------------|
| 1             |                                                  | 1 : 0.25                                       | 8                      |
| 2             | Positive stretching resistive effect             | 1 : 0.5                                        | 6.7                    |
| 3             | Negative stretching resistive effect             | 1 : 1                                          | 5                      |
| 4             |                                                  | 1 : 1.5                                        | 4                      |
| 5             |                                                  | 1 : 2                                          | 3.3                    |
| 6             | Positive stretching resistive effect             | 1 : 2.5                                        | 2.9                    |
| 7             |                                                  | 1 : 3                                          | 2.5                    |
| 8             |                                                  | 1 : 4                                          | 2                      |
| 9             |                                                  | 1 : 8                                          | 1.1                    |

Table S1. The ratio of CNT solution to NL solution with corresponding CNT weight ratio.
Table S2. The performance comparison of the conventional and our self-adaptive optogenetics devices.

|                        | Conventional optogenetics [37]                              | Self-adaptive optogenetics                      |
|------------------------|-------------------------------------------------------------|-------------------------------------------------|
| System integration     | Multi individual equipment                                 | An integrated device                            |
| Duty cycle             | 40% in both diastole and systole                           | 40% in diastole and 13% in systole              |
| Light radiance regulating | By individual regulator                              | Self-adaptive by negative stretching sensors    |
| Cure time              | 30 min                                                     | 8 min                                           |
| Electromagnetic interference | Distortion of the heart rate waveform                  | Steady heart rate recording                     |
Movie S1. Dynamic process of the cracks and wrinkles formation on the surface of negative stretching-resistive strain sensor during stretching.

Movie S2. Light intensity variation of an LED induced by the negative stretching-resistive strain sensor during stretching.

Movie S3. Closed-loop and self-adaptive cardiac optogenetics device based on negative stretching-resistive strain sensors and the enabled LED stimuli array in the ventricular tachycardia treatment.
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