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Letter

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Screen-Printed PEDOT:PSS Electrodes on Commercial Finished Textiles for Electrocardiography

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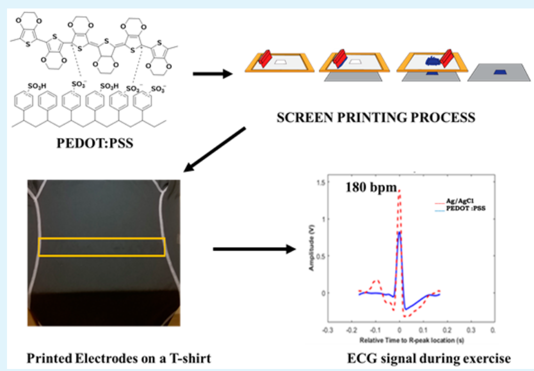
Supporting Information

ABSTRACT: Electrocardiography (ECG) is an essential technique for analyzing and monitoring cardiovascular physiological conditions such as arrhythmia. This article demonstrates the integration of screen-printed ECG circuitry from a commercially available conducting polymer, PEDOT:PSS, onto commercially available finished textiles. ECG signals were recorded in dry skin conditions due to the ability of PEDOT:PSS to function as both ionic and electronic conductors. The signal amplifies when the skin transpires water vapor or by applying a common lotion on the skin. Finally, PEDOT:PSS wires connected to PEDOT:PSS electrodes have been shown to record ECG signals comparable to Ag/AgCl connected to copper wires.



KEYWORDS: *PEDOT:PSS, screen printing, electrocardiogram, mixed conduction, wearable electronics*

Continuous health monitoring is an essential tool for accessing vital physiological functions. In 2012, cardiovascular diseases constituted about 42.8% of all noncommunicable deaths.[1] One of the most important techniques used for monitoring physiological function is electrocardiography (ECG). An electrocardiogram displays the change in magnitude of the cardiac vector and its direction as the heart pumps blood throughout the body. Typically, ECG electrodes are placed on the chest and limbs to measure the potential differences in the body that exist due to the varying electric field on the skin epidermis. Traditionally, Ag/AgCl electrodes are used for ECG monitoring, accompanied by a hydrogel containing KCl electrolyte that reduces the contact impedance between the skin and electrode. Additionally, adhesive around Ag/AgCl on the electrode helps establish better skin contact and lower motion artifacts.[2] However, Ag/AgCl has been shown to cause skin irritation during prolonged usage, and drying of the hydrogel leads to unreliable ECG data.[3]

Recent interest in measuring the physiological activity of the body from the epidermis has led researchers to explore nonconventional materials such as silver nanowires (AgNWs),[4] carbon nanotubes (CNTs),[5] graphene,[6] and carbon black (CB)[7] for recording the ECG signal. Hydrophobic PDMS conductive composites have made measurements of ECG possible in various underwater conditions.[5,7,8] Taji and co-workers[9] showed that minimizing skin contact impedance with the electrode plays an important role in the transduction of the ECG signal. Besides taking into account the material aspects of ECG measurement, the comfort and biocompatibility of the electrode should also be considered from the standpoint of wearable electronics.[10]

Poly(ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) (Figure 1(a)), a well-known conducting polymer, can be easily processed into films using traditional techniques such as inkjet printing, brush printing, and screen printing.[11] Recently, PEDOT:PSS has been explored for biopotential monitoring techniques, such as ECG,[12] EMG,[13] and EEG,[14] and has been successfully integrated into textiles.[12] PEDOT:PSS can be made more conductive by facilitating the phase segregation of PEDOT chains from PSS. This can be achieved by adding chemicals such as dimethyl sulfoxide (DMSO) (Figure 1(b)-1) and ethylene glycol, also known as secondary dopants.[15] In addition to DMSO, Triton X-100 (Figure 1(b)-2), a nonionic surfactant, helps the spreading of PEDOT:PSS, forming a continuous network structure.[16] The surface chemistry of the nanoparticles can also be explored to induce phase separation. Guo and co-workers explored the open air fabrication of PEDOT:PSS using roller printing, which led to specific current densities at the breakdown to be ca. 1600 A cm/g.[11,17] Nonwoven polyester fabric containing silica, as a

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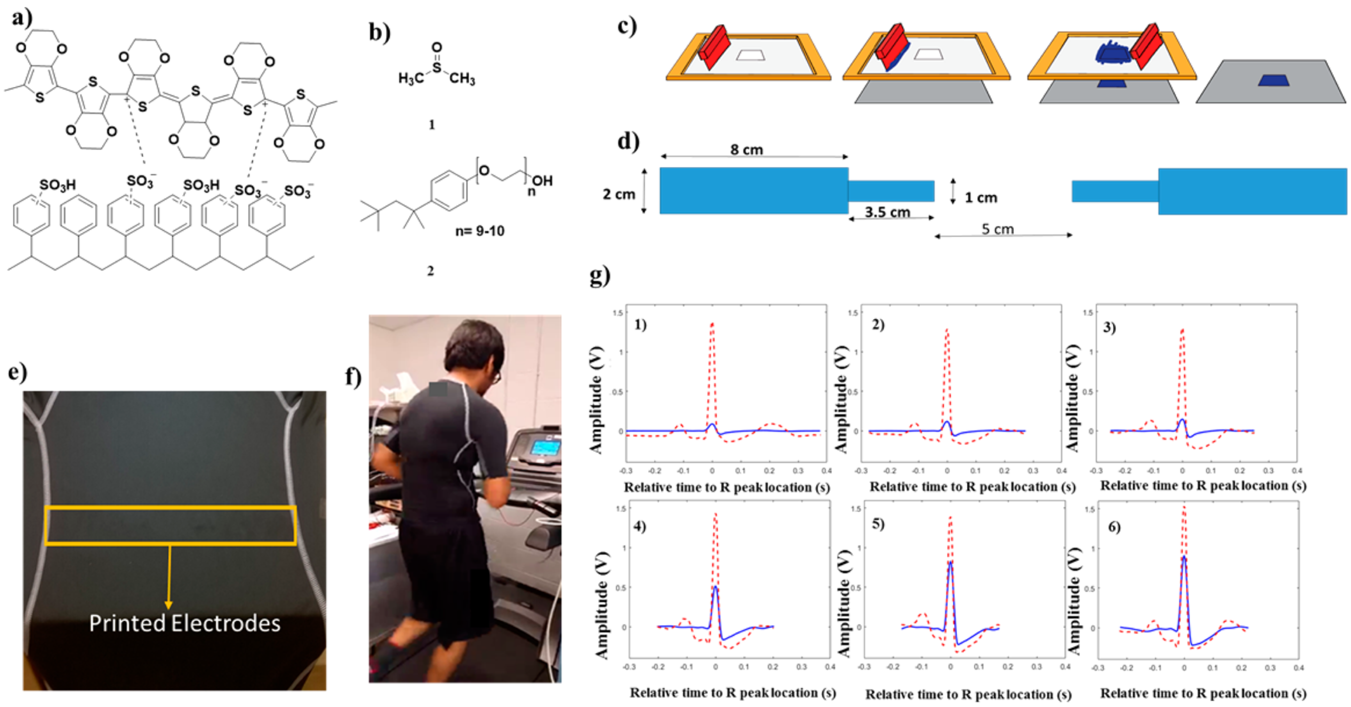

Figure 1. (a) Structure of PEDOT:PSS; (b) Additives of screen-printing formulation: (1) Dimethyl sulfoxide (DMSO) and (2) Triton X-100; (c) Screen-printing schematic; (d) The dimensions of the ECG electrodes; (e) PEDOT:PSS electrodes printed on a t-shirt; (f) Image of testing the t-shirt while exercising; and (g) ECG waveforms obtained from PEDOT:PSS electrodes on t-shirt (blue line) and Ag/AgCl electrodes (red dashed line) under different heart rates: (1) 80 bpm, (2) 110 bpm, (3) 140 bpm, (4) 150 bpm, (5) 180 bpm, and (6) 120 bpm.

delustering agent, when fully soaked with PEDOT:PSS has been shown to have high current carrying capacities of ca. 10 A/mm^2.[18] Considering that every three units in PEDOT carry a positive charge, which is stabilized by one unit of negative charge on PSS, calculations suggest that approximately 4 out of every 5 units of styrene sulfonic acid in polystyrenesulfonate are unbound (S2, Supporting Information). These unbound groups are available to react with the fabric through a covalent bond between the silanol groups contained in the silica and the free sulfonic acid groups of polystyrene sulfonic acid (PSSH). The pK_a value of sulfonic acid groups in PSS has been reported to be 1 and that of silanols in silica to be 6.8.[19,20] Pokrovsky and co-workers[21] studied the protonation of silanol groups and suggested the existence of $Si(OH_2)^+$ at an extremely low pH (0−3). Wet PEDOT:PSS on fabric will cause protonation of silanol groups, which, followed by condensation with sulfonic acid, leads to the formation of sulfo-silyl esters leading to the covalent bond formation with the fabric. Otley and co-workers proposed this reaction and its role in the phase separation of PEDOT:PSS, resulting in low sheet resistances of silica-containing fabrics coated with PEDOT:PSS.[18] The reaction of free sulfonic acid groups of PSS has later been suggested by Cho and co-workers.[22] This nanoparticle-induced phase separation leading to low sheet resistances can be used to make organic wires and biopotential electrodes, having current carrying capacities approaching copper. Other techniques that involve functionalization of textiles include grafting[23] and cross-linking[24] for self-cleaning, antimicrobial, and water capture properties.

In this paper, we describe the development of a conductive t-shirt for monitoring ECG signals using commercially available materials and screen-printing technique without the use of a hydrogel or an adhesive around the electrodes. Typically, commercial ECG electrodes consist of a Ag/AgCl electrode, a hydrogel consisting of an electrolyte, and an adhesive that helps in establishing skin contact. PEDOT:PSS is known to be a mixed conductor[25] in which PEDOT is responsible for electronic conductivity, and PSS contributes to ionic conductivity, both of which are necessary for transduction of the ECG signal. Screen-printed PEDOT:PSS electrodes on a t-shirt present an interesting material for monitoring the cardiovascular activity of an athlete, not only during sedentary conditions but also during exercise.

A viscous formulation of commercially available PEDOT:PSS (PH 1000), DMSO, and Triton X-100 was used for screen printing (Figure 1(c)). The solid content was obtained from thermogravimetric analysis (TGA) (Figure S1, Supporting Information). The design of the electrodes on the t-shirt is shown in Figure 1(d). One subject was recruited to test the viability of the electrode for measuring the ECG signal during exercise on a treadmill. An elastic chest strap was used to enhance skin contact with the electrodes. Ag/AgCl electrodes were used as a control to simultaneously measure ECG along with printed PEDOT:PSS electrodes. Figure 1(g) shows the ECG signal obtained from a subject under various exercising conditions. A discernible ECG signal was obtained with printed electrodes under dry skin conditions during the start of the exercise. The amplitude of this signal increased with an increase in the level of physical activity. When the subject was in recovery, the amplitude of the ECG signal remained the same (Figure 1(g)-6). The increase in the amplitude of the signal could be attributed to transpiration of water vapor from the skin during exercise leading to decrease in skin contact impedance. This observation is further substantiated by the results obtained from the electrochemical impedance spectroscopy, which shows that the electrode impedance of PEDOT:PSS fabric drops when the fabric is fully soaked in water (Figure S2, Supporting Information). This reduced

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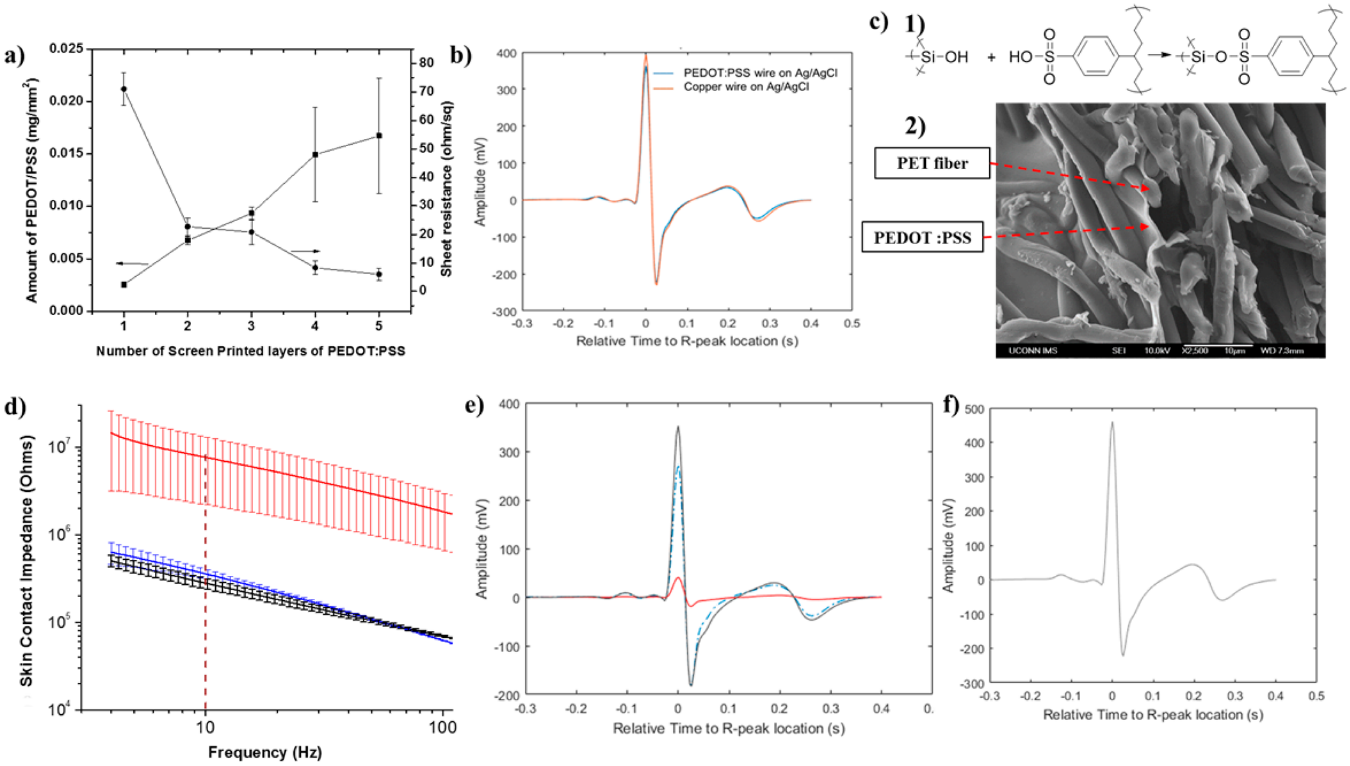

Figure 2. (a) Variation of sheet resistance and amount of PEDOT:PSS with an increasing number of screen-printed layers. (b) Overlay of ECG signals obtained from two different electrode-wire configurations with Ag/AgCl electrodes: PEDOT:PSS wire (blue line) and copper wire (orange line). (c) (1) Reaction scheme of the silanol group with the sulfonic acid of PSS to form silyl sulfonate ester and (2) SEM image of PEDOT:PSS-coated fabric electrode. (d) Variation in skin contact impedance of different electrodes: PEDOT:PSS electrode in dry conditions (red line), commercially available Ag/AgCl electrode (blue line), and PEDOT:PSS fabric electrode with lotion on skin (black). (e) ECG responses of different electrodes under different skin conditions from subject 1: PEDOT:PSS electrode in dry skin conditions (red line), PEDOT:PSS electrode with lotion on skin (black line), and Ag/AgCl (blue dash-dot line) at rest. (f) ECG response from a screen-printed PEDOT:PSS electrode with PEDOT:PSS wire with lotion on skin at rest.

contact impedance existed even after completion of exercise because of the hydrophilicity of PSS in PEDOT:PSS. The cross-correlation indices of ECG templates between the fabricated sensors and Ag/AgCl electrodes were approximately 83%, even in S4 and S5, where there are high motion artifacts. The average heart rate was calculated by measuring the R−R interval and was found to be as high as 180 bpm (Figure S3, Supporting Information) which corresponds to heart rate during high intensity interval training (HIIT). HIIT is generally done at a heart rate that is 85%−95% of the maximum heart rate (HR_{max}) which is calculated as HR_{max} = 207 − (0.7 × age),[26,27] and therefore the application of this type of shirt can be extended to almost any age group above 15 years. To demonstrate the reusability of the t-shirt, the ECG signal was recorded at a heart rate of 170 bpm after washing the t-shirt with commercially available detergent in a regular washing machine followed by hot air drying (Figure S4−S5, Supporting Information). Thus, PEDOT:PSS-based fabric electrodes are good candidates for measuring an ECG, and the applications could extend to gaming and military as well as sports medicine. To evaluate the skin compatibility of PEDOT:PSS, a preliminary study was carried out to check changes in pH of deionized water and artificial sweat solution when fabric swatches coated with PEDOT:PSS were immersed into solution. The pH of both the solutions changed to 3.17 from an initial value of 6.8 for water and 3.81 from 4.27 for artificial sweat at 25 °C (Figure S6, Supporting Information). The only conclusion that can be drawn from these initial studies is that protons can be lost from PEDOT:PSS coated on fabrics to water and/or sweat in contact with the fabric; however, the conditions tested here do not model conditions of a fabric in contact with skin. A more detailed study would be required to assess the toxicity of PEDOT−PSS.

To understand the material aspects of the wearable ECG sensor, a systematic study was carried out to replace components of a typical ECG setup, which currently includes copper wires and Ag/AgCl electrodes. An optimization study was done by screen printing PEDOT:PSS on nonwoven polyethylene terephthalate (PET) fabrics. PEDOT:PSS-coated textile was first demonstrated to function as a wire of 1 mm thickness with Ag/AgCl as an ECG electrode. To evaluate the change in resistance versus the amount of PEDOT:PSS on the fabric, a maximum of five layers of 2.5 cm length and 1 mm width were printed onto PET nonwoven textile. Figure 2(a) shows the change in sheet resistance of the fabric as a function of wire thickness. Five layers of PEDOT:PSS over an area of 25 mm^2 having a combined mass of ca. 45 mg gave a sheet resistance of 5.6 Ω/sq. To demonstrate the practical usage of the printed wires, a wire of length 10 cm and width 1 mm was printed onto PET. Three coatings of PEDOT:PSS were applied, and the overall resistance of the printed wires was measured to be ca. 3 kΩ. This wire was then connected to a Ag/AgCl electrode attached to the chest, and an ECG signal was recorded as shown in Figure 2(b). The change to PEDOT:PSS wires resulted in ca. 2% loss of amplitude of the

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ECG signal when compared to Ag/AgCl electrodes with copper wires of diameter 1 mm.

Screen-printed PEDOT:PSS fabrics were then evaluated as electrodes for ECG application. The ECG signals collected from a screen-printed PEDOT:PSS fabric that has an average resistance of ca. 5 Ω/sq were then compared with that of the commercially available Ag/AgCl electrode. To reduce the skin-electrode impedance, approximately 0.5 g of Cetaphil lotion was applied onto the skin as opposed to coating the electrode with a conducting gel. The skin contact impedances of electrodes at 10 Hz and the frequency that corresponds to the QRS complex were measured to be 0.35 ± 0.04 MΩ and 0.27 ± 0.6 MΩ for Ag/AgCl and screen-printed PEDOT:PSS electrodes with lotion, respectively. This decrease in contact impedance substantially affected the amplitude of the ECG signal with screen-printed electrodes, showing a 55.4% increase in amplitude compared to Ag/AgCl. Figure 2(d) shows the variation of skin contact impedance with frequency. Screen-printed PEDOT:PSS electrodes with underlying lotion on the skin have similar contact impedances to Ag/AgCl over a frequency range of 4−150 Hz. Thus, lowering the skin contact impedance plays a vital role in ECG recording, though a discernible ECG signal was still recorded with fabric PEDOT:PSS electrodes in dry skin conditions.

The contact impedance at 10 Hz was marginally lower for PEDOT:PSS electrodes with lotion on the skin than with dry electrodes, which further translates to variations in amplitude of ECG signal, as shown in Figure 2(e). Takamatsu and co-workers coated an ionic liquid gel on PEDOT:PSS electrodes on a knitted fabric and found that the impedance was 2.5 times lower than that of Ag/AgCl electrodes.[12] Pani and co-workers[28] matched the ECG signal obtained from a dip-coated PEDOT:PSS polyester electrode to Ag/AgCl by using a commercially available ECG gel on the skin and by wetting the electrode with saline. These results indicate that PEDOT:PSS acts as a transducer, even in dry conditions, and its performance can be improved by preparing the skin with lotion or by sweating.

The complications in ECG signals are caused by unwanted motion artifacts and noise. While the motion artifacts of a Ag/AgCl electrode have been alleviated by adhesive around the electrode, the motion artifacts of PEDOT:PSS-based fabric electrodes have not been fully explored. In this study, the conformity of the compression t-shirt onto the skin ensured that electrodes stayed in place during measurements which were further reinforced by a chest strap. The other factor which affects the quality of the ECG signal is the thermal noise, which is generated by the inherent resistive characteristic of PEDOT:PSS and is proportional to square root of its resistance for a given bandwidth.[29] The signal-to-noise ratio (SNR) of the PEDOT:PSS fabrics with a sheet resistance of 5 ohm/sq was found to be ca. 15.42 dB under dry skin conditions which increased to ca. 29.59 dB upon application of lotion on skin. Takamatsu and co-workers[12] reported the value as ca. 16.3 dB for their electrodes coated with a cross-linked gel and having a sheet resistance of 220 ohm/sq. Bihar and co-workers[30] found the SNR to be ca. 11.01 dB for three layers of inkjet-printed PEDOT:PSS on paper. This result points out how the resistance, in the form of thermal noise, together with skin contact impedance influences the signal quality. The signal becomes noisy with an increase in resistance of the electrode (Figure S7, Supporting Information). Additionally, the resistance of PEDOT:PSS-coated fabric is not affected by temperature from 0 to 100 °C.[17] The electrodes used in this study have a sheet resistance of ca. 5 Ω/sq and, therefore, can be used to measure ECG signal in real time conditions where the temperature could vary between 0 and 50 °C. Thus, screen-printing techniques combined with surface chemistry of fabrics can be exploited to coat PEDOT:PSS with low sheet resistances. Also, the method of screen printing is already established in the industry for making organic electronic devices, which would allow for scalability in the production of ECG electrodes.

PEDOT:PSS has not yet been used before as both wires and ECG electrodes. PEDOT:PSS electrodes were connected with PEDOT:PSS wires via snap buttons for recording ECG signals. Drawing on conclusions from the initial ECG testing, the skin was prepared by applying 0.5 g of lotion over an area on skin with length 40 cm and width 6 cm. The ECG waveform obtained is shown in Figure 2(f). The amplitude of the signal obtained was found to be comparable with that of Ag/AgCl. This result paves the way for organic materials to be integrated into wearables with the possible replacement of metals as both wires and electrodes.

This work describes the fabrication of an all organic ECG electrode using a commercially available conducting polymer onto commercially sold fabrics. Exploiting the mixed nature of conduction, PEDOT:PSS-based electrodes were shown to record ECG signal in dry condition. The formation of sulfo-silyl esters has been attributed to low sheet resistances of ca. 5 ohm/sq leading to low thermal noise. Screen-printed PEDOT:PSS electrodes on a t-shirt were shown to record ECG signal under physical activity at a heart rate of 180 bpm. Increased ionic conductivity brought about by transpiration of water results in an increase in the amplitude of the signal. PEDOT:PSS demonstrated the ease of processing in that it can be used as an organic wire as well as an electrode, resulting in a signal comparable to Ag/AgCl electrode. By integrating these electrodes, wires, and potentially more complex circuitry directly into fabrics, smart clothing could be developed for continuous monitoring in both athletes and patients having cardiovascular risk, increasing their quality of life.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsami.7b09954.

Experimental details and procedures, fabrication of ECG electrodes, and measurement of ECG signal (PDF)

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Notes

The authors declare no competing financial interest.

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