Plant-Based Biodegradable Capacitive Tactile Pressure Sensor Using Flexible and Transparent Leaf Skeletons as Electrodes and Flower Petal as Dielectric Layer

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In biomedical sciences, there is demand for electronic skins with highly sensitive tactile sensors, having applications in patient monitoring, human–machine interfaces, and on-body sensors. In clinical applications, it would be especially beneficial if the sensors would be disposable. Here, an all plant-material-based biodegradable capacitive tactile pressure sensor for disposable electronic skins is reported. Silver-nanowire-coated leaf skeletons are used as breathable and flexible electrodes while freeze-dried rose petals are used as the dielectric layer. The leaf skeleton electrodes have a rough fractal-like architecture, which provides good adhesion to the silver nanowires and maintains interconnections between the silver nanowires when the electrodes are bent. The electrodes display low constant resistance up to curvature of 800 m⁻¹. The rose petal dielectric layer has a multiscale 3D cell wall microstructure, which compresses elastically when subjected to pressure. The fabricated sensor can respond to pressures ranging from 0.007 to at least 60 kPa, with a maximum sensitivity of −0.08 kPa⁻¹. The signal is stable for at least 5000 pressure cycles, after an initial break-in period. Owing to the all biomaterial constituents, the sensor is biodegradable under aqueous conditions. The sensor is successfully applied as an e-skin in touch sensing and gesture monitoring.

Electronic skins (e-skins) with multiple functions attract interest due to their promising applications in patient monitoring, skin prosthetics, soft robotics, human–machine interaction, and wearable electronics.[1–7] To achieve high-precision on-body sensing or practical healthcare monitoring, an ideal e-skin should be highly flexible, sensitive, low-cost, facile to fabricate, light-weight, and breathable. Furthermore, clinical applications would benefit from the sensor being disposable and easily recyclable, to have the sensors sterile and to limit the growing amount of electronic waste (e-waste).[8,9] For recyclability, it would be highly beneficial if electronic waste could be degraded and decomposed, like most materials from nature.

E-skins based on capacitive,[10] piezoresistive,[11,12] piezoelectric,[13] and triboelectric[14] sensing mechanisms have been demonstrated. Capacitive e-skins are particularly promising because they have a simple design, high sensitivity, fast response up to the limit of viscoelasticity, high accuracy, less drift, and very low limit of detection.[3,15,16] Typically, the capacitive e-skis comprise of two parts: a dielectric layer, sandwiched between flexible electrodes. When the sandwiched structure is compressed, the capacitance changes, because the distance between the electrodes changes. Bulk elastomer, microstructured elastomer[15,16] or foams,[1] and polymer networks[17] have been used as the dielectric layer. To improve the sensitivity and the dynamic performance of the sensor, engineered microstructures—such as pyramids,[15] spheres,[18] or micropores[19]—are added to the dielectric layer using specialized microfabrication techniques. With such structures, the sensors display repeatable pressure response and do not get saturated at high pressures; however, efforts are being made in this field to cut the fabrication costs and to fabricate the highly sensitive dielectric layers which are eco-friendly.[20,21]

The other component in the e-skin is the electrode which can have conformal contact with the human body and should tolerate recurrent bending, twisting, and other deformations.[22] Many types of flexible electrodes have been developed for e-skins, including filamentary serpentine[23,24] and micro/nano-mesh-like[25] geometries. Still, there remain some major challenges for the development of electrodes which are permeable, nature-friendly, and allow optical transmission.[26–28] Designs based on kirigami (the art of paper cutting) have been used in the fabrication of flexible, permeable, and translucent/transparent electrodes,[29] but again required elaborate microfabrication. The complex fabrication procedures and use of harsh chemicals in most of the reported electrodes are some of the other issues which need to be addressed.
In this article, we report a facile method for fabricating capacitive e-skins out of plant-based materials. A leaf skeleton, coated with silver nanowires (Ag NW), was used as a breathable and flexible electrode while a rose petal was used as the dielectric layer. The electrode was fabricated by dispersing Ag NW on the leaf skeleton. The skeleton displayed a fractal network-like microstructure, with lignin-rich fragments interconnecting each other. The interconnected fractal design displayed by the leaf skeleton is advantageous here because, for engineering structures, these are known to provide better stability, optimal surface coverage, and facilitate the efficient distribution of thermal and electrical energy. Due to its network-like architecture, the electrode was transparent (≈60–75%) at wavelengths ranging from 400 to 900 nm. The electrode had a very low sheet resistance, ranging from 975 down to ≈4 Ω sq⁻¹, depending on the Ag NW loading. The electrode had a constant resistance even at a curvature up to 800 m⁻¹. Leaf skeleton electrodes have a rough fractal-like architecture, which provides good adhesion to the silver nanowires and maintains interconnections between the silver nanowires when the electrodes are bent. A rose petal was used as the dielectric layer. The petal displayed hierarchical microstructures and a 3D cell wall network. The rose petals were subjected to freeze-drying, which removes all water content from them. This improves the stability and the shelf life of the petals while maintaining the surface morphology and inner structural arrangements. These features act as a compressible architecture which elastically collapses under pressure, which contributes to the sensitivity of the sensor. The fabricated sensor can respond to pressures ranging from 0.007 to at least 60 kPa, with a maximum sensitivity of ≈0.08 kPa⁻¹. When subjected to 5000 pressure cycles, the sensor showed some small changes during the first few thousand cycles of loading, but after this initial break-in period, the signal was very stable. The noise level of the sensor was 0.003 pF which corresponds to reported lower limit of 0.007 kPa, by considering the approximated model for the sensor response in the low-pressure region. Owing to all biomaterial constituents, the sensor was also readily biodegradable under aqueous conditions. We also show that the fabricated capacitive sensor can be used as an e-skin for touch sensing and gesture monitoring. We believe that our strategy of directly using natural materials for the fabrication of capacitive sensors will put forward a facile, cost-effective, eco-friendly, and scalable approach to encourage flexible electronics.

The fabrication procedure of the sensor is outlined in Figure 1. To fabricate the leaf-based electrode, a leaf skeleton of a Bodhi tree (Ficus religiosa) was used as it shows fractal-like

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**Figure 1.** Fabrication of the plant-based capacitive tactile pressure sensor. a) A leaf skeleton is loaded with Ag NW. b) A freeze-dried rose petal serves as the dielectric layer. c) A capacitive tactile pressure sensor is made by sandwiching a dielectric rose petal layer between two leaf electrodes, with the sealing provided by cellulose tape. d,e) Scanning electron microscopy images of a leaf skeleton electrode and a freeze-dried rose petal, respectively. Scale bar in the insets: 1 μm. f) Scanning electron microscopy image displaying a cross section of the sensor. The hollow cavities inside the rose petals are evident.
interconnections of veins at the microscale and does not snap even after repeated bendings. It is mainly composed of a biopolymer that is rich in lignin and is responsible for mechanical strength.\(^{[30,33]}\) First, the surface of the skeleton was treated with hexadecyltrimethylammonium bromide solution to induce a partial positive charge on the surface. Then the leaf skeleton was treated with the Ag NW solutions of different concentrations (in ethanol) in the hydrophobic Petri plates until all the Ag NW solution gets soaked by the leaf skeleton (details in the Supporting Information). After drying, a leaf skeleton electrode was obtained which was slightly greyish in color and transparent. The 3D scaffold composed of lignin-rich biomaterials acts as a backbone and the highly conductive Ag NW adhered to the skeleton form a conductive network across the leaf skeleton. For the dielectric layer, rose petals were initially treated with 2% glutaraldehyde solution to fix the morphological features. This was followed by freeze-drying treatment to remove the water content and to preserve the outer and inner microstructures. This prevents curling of the surface due to dehydration. The simple assembly of the flexible capacitive e-skin is shown in Figure 1c. The capacitive e-skin consists freeze-dried rose petal dielectric layer sandwiched between two leaf skeleton electrodes. The edges were finally sealed together with biodegradable cellulose tape. The detailed fabrication process is given in the Supporting Information.

The scanning electron microscopy image of the leaf skeleton without any Ag NW treatment is shown in Figure S1a (Supporting Information). The structural network of veins of the leaf skeleton contains bundles of vessels and interconnected fibers can be clearly seen in Figure S1b (Supporting Information). These vascular bundles and fibers are particularly suited as a scaffold because they are rich in lignin-based biomaterials and provide mechanical strength.\(^{[30]}\) The overall surface displays a fractal-like appearance with the typical gap between the interconnections of fibers ranging from 100 to 400 µm. Figure 1d displays scanning electron microscopy image of the leaf skeleton electrode. The Ag NWs have an average aspect ratio of \(\approx 1000\). The nanowires are tightly adhered to the surface and are tangled together to form a continuous nanonetwork on the top of the skeleton fibers. Figure 1e displays scanning electron microscopy images of the rose petal surface (upper epidermis). The surface has hierarchical structures of micropapillae arrays with sub-micrometer folds. These folds are spread throughout the surface with an average distance of 1 µm between them. The cross-sectional images (Figure 1f) show that the epidermis layers and the hollow cavities composed of mesophyll cells.

The conductivity of the leaf electrode is dependent on the loading quantity of the nanowires. It is clear from Figure 2a that the sheet resistance of the leaf electrode drops significantly as the loading quantity of the Ag NW is increased. The corresponding surface morphologies are shown in Figure S2 (Supporting Information). The electrode displayed an average sheet resistance of \(\approx 980 \, \Omega \, \text{sq}^{-1}\) when initially loaded with 12.5 µg cm\(^{-2}\) Ag NW. The sheet resistance reaches as low as 4.3 Ω sq\(^{-1}\) when the loading quantity of Ag NW is increased to 200 µg cm\(^{-2}\), which is at par with the current state of the art.\(^{[22]}\) Thus, the conductivity of the leaf skeleton-based electrode can be adjusted by changing the loading quantity of the Ag NW. Along with its hexadecyltrimethylammonium bromide treatment ensures that the Ag NW is evenly spread and properly adhered to the surface.

Electrodes for e-skins should be flexible, without plastic deformation or cracking. To demonstrate flexibility, the leaf electrode was clamped from its edges and bent with the help of a linear translation stage, while recording the curvature of the leaf and the resistance of the electrode. The results are shown in Figure 2b. The resistance does not change much as the leaf is bent: the resistance increases from 120 only to 182 Ω as the curvature is increased from 0 to 800 m\(^{-1}\). In most of the scaffold-based electrodes, there is some variation in the resistance values during bending of the electrode which is due to the increased contacts between the Ag NW.\(^{[34]}\)

To see how the resistance of the leaf electrode changes with temperature, we measured the resistance values at different temperatures as seen in the inset of Figure 2c. At temperatures <180 °C, the resistance of the electrode slightly increased with temperature. Above 180 °C, the resistance of the leaf skeleton electrode increases substantially, which can be credited to the surface oxidation and snapping of the Ag NW due to the high temperatures.\(^{[22,28]}\) To show the aging of the leaf skeleton electrode, we kept the electrode in the ambient conditions for 58 d and measured its resistance. In 58 d, the sheet resistance of the electrode increased from 44 only to 80 Ω sq\(^{-1}\) (Figure S3, Supporting Information).

To show the transparency of the leaf skeleton electrode, we measured the transmittance spectrum at different areas in the skeleton electrode. Figure 2d shows the overall relative transmittance spectra of the leaf skeleton electrode at visible and near-infrared wavelengths. In the inner areas without any veins, the average relative transmittance at wavelengths ranging from 400 to 900 nm was found to be \(\approx 75\%\) while the areas with veins displayed average relative transmittance \(\approx 60\%\). In conclusion, the results in Figure 2 and Figure S3 (Supporting Information) validate the good performance of the leaf skeleton-based electrode.

To make a capacitive pressure sensor out of the electrode, leaf electrodes (Ag NW loading of 200 µg cm\(^{-2}\)) and a rose petal were layered on top of each other (Figure 1c). When pressure is applied, the capacitance between the electrodes varies and thus can be measured to quantify the pressure. We tested the sensor with a broad range of pressures from 0.1 to 60 kPa, which is close to the range of pressures experienced by human skin during natural activities.\(^{[35]}\) We used a mechanical tester with a 10 mm diameter cylindrical probe to gradually increase the pressure on the sensor. The load on the sensor was varied step-wise, while the capacitance was measured using an LCR meter. Between each experiment, the load was completely removed. Figure 3a shows the recorded capacitance change over time, and Figure 3b shows the same data plotted over pressure. In the low-pressure regime (<1 kPa), seven points were recorded to get an accurate estimation of the linearity of the sensor in this regime. Again using the same data, Figure 3c shows the sensitivity of the sensor as a function of pressure. The sensitivity \(S\) of the sensor was calculated according to the following formula:\(^{[30]}\)

\[
S = \frac{d(C/C_0)}{dP}
\]
where \( C \) is the capacitance, \( C_0 \) is the initial capacitance, and \( P \) is the applied pressure. It was observed that the sensitivity tends to decrease with increasing pressure, i.e., the capacitance increased sublinearly with pressure. Such a sublinear response is advantageous: the sensor has a high dynamic range. The highest sensitivity in the low-pressure range had
Figure 3. Characterization of the plant-based capacitive tactile pressure sensor. a) $C - C_0$ when the pressure is varied on the sensor. b) Data from (a), with relative capacitance, plotted as a function of the pressure. Inset: close-up of the low-pressure regime. c) Sensitivity, calculated using Equation (1) and plotted as a function of the applied pressure. d) Capacitance–pressure relation of a fresh sensor and a sensor after one month of storing in ambient conditions. e) Stability of the e-skin when dynamically loaded for 5000 cycles with a maximum pressure of 20 kPa. f) Recovery of the e-skin when the load of 12.5 kPa is removed after 15 min. g) Schematic of the compression of the dielectric layer. The air pockets in the dielectric layer collapse during the compression.
a maximum value of $\approx 0.08 \text{ kPa}^{-1}$, which is comparable to the sensitivity reported in the similar capacitive sensors integrated as e-skins.[20] To observe potential hysteresis in the sensor, we also measured the capacitance while ramping up and ramping down the pressure (Figure S4, Supporting Information).

To test how the sensor ages, the sensitivity was characterized after storing the sensor in ambient conditions for one month. The results were compared with those of a freshly fabricated sensor, as shown in Figure 3d. From the figure, it is evident that there is no substantial change in the sensor response; however, the initial capacitance of the sensor increased slightly with aging. This may be because of the collapse of cell wall structures of rose petals over time due to natural degradation.[36] This may lead to a decrease in the thickness of the dielectric layer which brings the electrodes closer to each other. In addition, the slight decrease in the performance of the sensor over time may be due to the Ag NW oxidation with time. Oxide formation is very common on the Ag surfaces in ambient conditions.[37] The oxide formation also leads to an increase in sheet resistance of surfaces after a few days which is evident in Figure S3 (Supporting Information).

To evaluate the dynamic characteristics of the sensor, the probe of the mechanical tester was moved up and down sinusoidally. We applied a cyclic pressure on the sensor for 5000 cycles with a frequency of 0.2 Hz, with a maximum pressure of 20 kPa. The results are shown in Figure 3e. The sensor showed some small changes during the first few thousand cycles of loading, but after this initial break-in period, the signal was very stable (Figure 3e, see also Figure S5, Supporting Information). This may be due to a few microstructures in the freeze-dried rose petals deforming plastically during the initial pressure cycles. This plastic deformation during initial pressure cycles can also be seen in the capacitive sensors utilizing fabrics and polymers.[38] Also, as the morphology of the leaf skeleton and rose petals is not perfectly planar, the interface between tape, electrode, and the rose petals takes some pressing cycles to settle which may be the cause of change in the capacitance. After a thousand cycles, the contact between the different layers becomes uniform to provide a stable signal which can be seen in Figure S5 (Supporting Information). To observe other dynamic characteristics such as drift and recovery time, the sensor was loaded initially with 12.5 kPa for 15 min, followed by quick removal of the load, after which the capacitance was recorded for another 10 min (Figure 3f). Compared to the results obtained from using a microstructured gel polymer as a dielectric layer,[23] recovery time is slightly longer and may be an issue in applications demanding high absolute accuracy. However, in many applications, we are only interested in the dynamic (high frequency) response, so the slow recovery may be a nonissue. In such applications, other merits—such as the ease of fabrication, the simplicity of structure, and biodegradability of the sensor—may be more important.

The sensing mechanism of the e-skin fabricated in this study is shown in Figure 3g. The sensor is essentially a plate capacitor, with capacitance $C$ directly proportional to the area $A$ of the electrodes, directly proportional to the permittivity $\varepsilon$ of the dielectric layer, and inversely proportional to the distance $d$ between the electrodes, i.e., $C = \frac{A\varepsilon}{d}$. Any variation in $A$, $\varepsilon$, or $d$ will lead to a variation in capacitance. For our sensors, $A = 2 \text{ cm}^2$ and $d = 130 - 200 \mu\text{m}$ (Figure 1f). When there is no load, the specific capacitance $C/A$ is less than $3 \text{ pF cm}^{-2}$ which increases up to 3.3 times at a load of 45 kPa. The cross-sectional images of the dried rose petal (Figure 1f) show that the dielectric layer consists of freeze-dried cell walls presenting hollow cavity-like structures with air trapped in between. This porous architecture makes the sensor compressible. When the pressure is applied, $d$ decreases mainly from the compression of the hollow cavities along with the deformation of the surface micro/nanofolds as seen in Figure 1e. This leads to very high sensitivity even at very low pressures. Here, freeze-drying of rose petals offers an advantage over naturally drying by preventing the collapse of the cavities and a significant decrease in the thickness and compliance of the dielectric layer. Due to the presence of air in the rose petals, it is useful to think of the sensor as two capacitors in series, i.e., $C = \frac{1}{1/(C_{\text{air}} + 1/C_{\text{petal}})} = \frac{A}{(d_{\text{air}}/\varepsilon_{\text{air}} + d_{\text{petal}}/\varepsilon_{\text{petal}})}$. Due to the collapse of the air pockets, it is safe to assume that the variation in $d_{\text{air}}$ is much larger than the variation in $d_{\text{petal}}$. On the other hand, the relative permittivity of dry leaves is usually $\approx 2$.[19] Therefore, most of the variation in the capacitance is from the variation in $d_{\text{air}}$, explaining why the elastically collapsible air pockets increase the sensitivity of the sensor.

One of the disadvantages of using leaf skeletons of a F. religiosa as the electrode substrate is that the leaf skeletons only flexible, not stretchable. To show this, a piece of leaf electrode (10 mm $\times$ 20 mm) was stretched linearly by the mechanical tester until it fractured. When the leaf fractured, the strain was 1% and the force was 2.7 N.

We demonstrated practical applications of the sensors by mounting them onto a robotic hand and onto a glove for monitoring human gestures (Figure 4). First, we placed the sensor at the fingertip of a 3D-printed robotic hand. The sensor can detect when a human finger touches the robotic hand repeatedly (Figure 4a, see also Figure S6, Supporting Information). Second, we integrated a sensor into each of the five fingers of a nitrile glove to convert it into a smart glove for gesture monitoring. Different gestures were performed in a sequence where each gesture was held for 15 s. The signal from each sensor clearly reflects the gestures of each finger, as shown in Figure 4b. No delamination of the layers and failure of the device was observed during the gesture monitoring experiments. These demonstrations show that the sensor can be used in practical applications.

To show that the sensors are biodegradable, we stored the sensor in tap water and phosphate-buffered saline solution and monitored the biodegradability for 75 d (Figure 5). After the experiment, the sensor showed a significant degradation in tap water, but less so in the buffer solution. We conclude that the sensor is biodegradable and can be processed easily as waste. In conclusion, we have used Ag NW decorated leaf skeletons as electrodes and freeze-dried rose petals as a dielectric layer to fabricate highly sensitive capacitive tactile pressure sensors. Being biodegradable and entirely made of plant-based materials, these sensors may help to reduce the carbon footprints and e-waste. It should be pointed out that the leaf electrode is highly flexible, but not stretchable. Nevertheless, the leaf skeletons of F. religiosa used in this study are not the only natural materials that can be used as the electrode materials. There
are many unexplored biological surfaces—skeletons of other leaves and plant-based membranes—that may provide interesting interconnected architecture at the microscale, good stretchability, good adhesion to conducting nanomaterials while maintaining optical transparency and breathability. In addition, there may be many biological surfaces belonging to different plant species displaying 3D micro/nanofoam-like architecture so they could be used as soft dielectric layers for high-sensitivity...
capacitive sensors. The natural materials we used for the fabrication of the e-skin are environment-friendly, low cost, readily available, and facile compared to artificial microstructured surfaces fabricated by expensive and complex techniques using plenty of harsh chemicals.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

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