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3D Printed Stretchable Tactile Sensors

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The development of methods for the direct three-dimensional (3D) printing of multi-functional stretchable electronic devices in various form factors and on a variety of surfaces could impact areas ranging from wearable electronics, energy harvesting devices, and prosthetic bionic skins, to fundamental studies on soft mechanics and biotic-abiotic interfaces. In the past decade, the development of stretchable electronic devices and sensors has dramatically accelerated, concomitant with advances in functional materials and fabrication processes. In particular, novel strategies have been developed to enable the intimate biointegration of wearable electronic devices with human skin in ways that bypass the mechanical and thermal restrictions of traditional microfabrication technologies.^[1–2] In developing wearable electronics, several factors are of paramount importance to consider, including mechanical flexibility, device scalability, fundamental interfacial mechanics and chemistries, and retaining high performance of the functional materials and devices.^[3] Mechanical flexibility is particularly important for devices that are in direct contact with certain areas of the skin (Young's modulus ~ 130–657 kPa),^[4] in order to minimize discomfort and respond to the various strain degrees of freedom associated with body motions. One important strategy for achieving high stretchability in wearable electronic devices is to modify the form factors in structures created from established functional materials, devices, and fabrication techniques. For example, ultrathin silicon or gold films patterned onto pre-strained substrates, followed by strain release, form into buckled geometries which permit stretchability when strained in two and three dimensions, including deformations on 3D curved structures.^[5–6] Other routes toward assembling stretchable devices include: (i) utilizing stretchable polymer composites with conductive fillers,^[7–8] and (ii) employing thin conductive films on stretchable polymer substrates.^[9–10] These strategies are typically based on cutting-edge adaptations of conventional 2D planar fabrication processes, such as spin coating, photolithography, and metal deposition, and thus require expensive cleanrooms to enable their implementation. This potentially restricts the adaptation of such techniques to in-the-field, off-grid operations where devices are desired on demand from only raw materials.

As skin plays a vital role in shaping our interactions with the world, stretchable electronic devices mimicking the properties of skin could have profound implications for prosthetics and medicine.^[11–12] Providing prosthetic hands with the ability to sense is particularly important, as each year ca. 185,000 Americans suffer from limb loss, requiring substitutional prosthetics.^[13] Efforts to create artificial skins with human-like sensory capabilities have been motivated over the past several decades.^[14] Common methods to measure tactile stimuli are based on capacitive and piezoresistive mechanisms. Capacitive sensors can exhibit excellent sensitivity, linearity, and temperature invariance, but their performance can be influenced by electromagnetic interference, often requiring shielding to achieve high signal-to-noise ratios.^[15–16] Piezoresistive sensors have been extensively investigated in artificial skin applications owing to their flexibility, simple device structure, and compatibility with electronic readout methods.^[14] New methods to fabricate piezoresistive artificial skins directly onto conformal, geometrically complex structures could further expand their impact.

3D printing technologies typically employ a computer-controlled positioning stage whose motion is derived from a prescribed computer-generated pattern in order to directly create desired 3D micro- and macrostructures from raw materials and inks, including the ability to construct complex, customized 3D geometries.^[17–20] Recent innovations in 3D printing also offer flexibility in material selection,^[18, 21] including cells,^[22–25] semiconductors,^[26] metals,^[27] ceramics,^[28–29] and polymers.^[30–31] Hence, 3D printing has resulted in the manufacturing of a variety of functional materials & devices, including soft sensors,^[32–35] electronics,^[26, 36] biomedical devices^[37–39] and artificial tissues and organs.^[23–25] In addition, recent developments in computer modeling, ink compositions, and extrusion-based deposition techniques have enabled (i) multi-material 3D printing techniques which allow for the fabrication of 3D objects with higher levels of complexity and functional performance,^[40–42] and (ii) the combination of 3D imaging technologies and 3D printing for next-generation, patient-specific biomanufacturing initiatives.^[23] Together, these multifaceted advanced manufacturing techniques provide new opportunities for the fabrication of wearable devices via the incorporation of topological conformalities, interwoven 3D geometries, and the programmable integration of multiple materials and functionalities.^[37] Indeed, directly printing 3D structures and functional devices on biological and non-flat substrates remains a substantial challenge which requires, at least, closed-loop scan-and-print processes, biocompatible inks which can be processed under mild conditions, and a fundamental understanding of interfacial chemistries and mechanics.

Here, we demonstrate the design and fabrication of stretchable tactile sensors which are 3D printed under ambient conditions via a combination of nanocomposite ink optimization, 3D imaging, and multi-material 3D printing. For this purpose, we developed sinter-free inks with adjustable viscosities and electrical conductivities, integrated these materials via the same one-pot custom-devised 3D printing process, and measured their sensing behavior on various platforms and in fully integrated arrays. The developed inks were manipulated by four different, independently-addressable nozzles in order to fabricate the 3D tactile sensors via one continuous 3D printing process. Each printed device was composed of a base layer, a sensor layer, two electrode layers, an isolating layer, and a sacrificial supporting layer (Figure 1a–c). The complete step-by-step process is shown in Figure 1d.

A critical aspect of 3D printing wearable devices is to enable printing under mild, ambient conditions, compatible with various biological substrates. In order to accomplish this, the ink materials should be optimized in order to (i) ensure smooth flow through fine deposition nozzles during the printing process, (ii) promote high stretchability while maintaining good electrical conductivity of the printed features, but without curing or sintering in harsh conditions (i.e., high temperature, intense UV), and (iii) provide sufficient structural integrity to withstand drying without delamination or distortion. Initially, we explored the incorporation of nanoscale conductive fillers such as silver (Ag) nanoparticles (Figure S1a) and nanowires (Figure S1b) in the stretchable polymers to achieve both high conductivity and stretchability. However, these nanomaterials required high temperature sintering to form a continuous conductive network. Conversely, microscale particles such as silver flakes were added into an elastomeric polymer to fabricate elastic conductors in a stencil printing step (Figure S1c).^[8] However, their resolutions were limited due to the aggregation of these large-sized particles, which obstructed ink flow through a fine nozzle.

To overcome these limitations, here we developed a new ink enabled by mixing submicron-sized silver particles within a highly stretchable silicone elastomer (Dragon Skin® 10,^[43] which is curable at room temperature). Figures 2a–b show the typical uniform particle distributions of two inks with 68 wt% and 75 wt% Ag, respectively, incorporated in the silicone elastomer. The rheological properties and pot-life of the inks were adjusted by adding chemical agents including a curing retarder, a thickening agent, and a solvent (dichloromethane (DCM)). With increasing Ag content, the stretchability of the cured inks decreased while the modulus increased (Figure 2c–d, and Figure S2).

To identify the optimal sensor layer ink, the electrical properties of the cured inks were analyzed via four-point probe measurements. The electrical conductivities of the Ag/silicone composites showed typical Ag particle concentration-dependent behavior. The conductivity increased with loading of silver particles, and the percolation threshold was found to be 67.45 wt% (Figure S3a). Thus, near the percolation concentration (68 wt%), the three dimensional conductive network is not fully formed,^[44] such that the conductivity of the ink with 68 wt% Ag loading content should be sensitive to applied pressure. Indeed, when a continuous pressure was applied to the ink with 68 wt% Ag, its resistance dramatically decreased and finally reached a plateau (Figure 2e), resulting in a ca. 200× change in resistance. Under compression, Ag particles are forced to pack more closely, increasing electrical conduction pathways,^[45] and the ink containing 68 wt% Ag concentration was the most sensitive elastic piezoresistive material among our prepared inks.

When the 68 wt% Ag ink was tested as a sensor layer, the relative current changes (I/I_0) were found to be 2500%, 8500%, and 17000% under 60, 120, and 250 kPa cyclic applied loading pressures, respectively (Figure 2f). In addition, the much lower initial conductivity ($9.9 \times 10^{-3} \text{ S m}^{-1}$) and high sensitivity of the inks with 68 wt% Ag content ensured that the primary electrical signal of the final device arose from deformation of the sensor layer. As the Ag content increased to 75 wt% or higher, the conductive network inside the inks surpassed the percolation threshold, and the relative current change decreased to 900% (for 75 wt%) and 280% (for 80 wt%), when an equivalent cyclic pressure of 250 kPa was applied

(Figure S3b–c). Thus, the ink with 75 wt% Ag was chosen for the electrode layers due to its appropriate balance of high conductivity and stretchability.

Besides the sensor and electrode layers, the base and isolating layers were fabricated using a modified silicone ink, Dragon Skin® 10, with both a thickening agent and curing retarder added. After printing and curing the ink at room temperature, the high stretchability of Dragon Skin® 10 (~1000% strain to failure)^[43, 46] offered the tactile sensor both mechanical flexibility and electrical isolation. Further, the silicone rubber has a low modulus of 150 kPa,^[46] similar to that of skin, and lower hardness (Shore A 10) than that of human skin (Shore A 20).^[47–48] Based on the above analysis of the inks, our tactile sensors were fabricated as follows (Figure 1d and Movie S1): (i) Base layer: silicone, (ii) Bottom electrode: 75 wt% Ag/silicone, (iii) Sensor layer: 68 wt% Ag/silicone, (iv) Isolating layer: silicone, (v) Supporting layer: 40% Pluronic (removed later, see Movie S2), and (vi) Top electrode: 75 wt% Ag/silicone (Figure S4). The final printed devices are shown in Figure 3a–c.

To measure the mechanical properties of the devices, we conducted dynamic mechanical analyses (DMA) on the printed devices to determine their mechanical responses under dynamic applied forces. As shown in Figure S5, both the storage (E') and loss (E'') moduli increased two orders of magnitude as the dynamic strains increased from 10% to 50%. On the other hand, the loss modulus slightly increased by a factor of 2 at higher dynamic frequencies, which is typical viscoelastic behavior.^[49] We also conducted a finite element analysis (FEA) of the printed tactile sensor under compressive loading conditions. A simplified 3D tactile sensor model was created and meshed (Figure S6), then an Ogden model was found to accurately fit the hyperelastic properties of each component (Figure S7).^[50] Hence, the Ogden model was employed to simulate the properties of the printed device, with results shown in Figure S8. The regions with higher stress and strain concentration were primarily located in the sensor layer upon typical loading scenarios, which indicated that the mechanical response of the device under compression was dominated by the properties of the 68 wt% Ag/silicone ink. By contrast, when the device was cyclically stretched to 200% strain under a tensile stress, the deformation was generated on the electrode layers, and no obvious changes were observed on the sensor layer (Figure S9 and Movie S3). This result revealed that the components with 75 wt% Ag/silicone ink determined the mechanical properties of the device under stretching.

We next evaluated the electrical sensing behavior of the printed tactile sensors. When the top and bottom electrodes were contacted, the device revealed linear I – V curves (Figure 3d). In addition, as the applied pressure increased from 100 to 500 kPa, the resistance of the device decreased ~12-fold, from 1.14 k Ω to 95 Ω . Under dynamic testing, the amplitude of output current signals significantly decreased as the input frequency increased from 0.125 to 2 Hz (Figure 3e–f and Figure S10). A negligible hysteresis was observed when the frequency was below 0.25 Hz, and the relative hysteresis increased with frequency up to 82% at 1.0 Hz. The hysteresis can be attributed to the elastic deformation of the device during loading-unloading cycles, and the viscoelastic behavior of the silicone.^[51]

The sensitivity of the tactile sensor is defined as the compressive gauge factor (GF) and calculated using Equation S16 (Supporting Information). For the entire device it was found to be ~ 180 (Figure 3g), which was similar to that of the 68 wt% Ag/silicone ink, to single crystal silicon, and to other recent studies.^[51–52] This phenomenon demonstrated that the sensor layer (made with the 68 wt% Ag/silicone ink) dominated the electromechanical properties of the printed devices under compressive strains. Moreover, when a constant strain (initial pressure 500 kPa) was applied to the device, the current change sharply increased by 8000% within a few seconds; and the output signal gradually reached a plateau. The applied pressure exhibited relaxation behavior (Figure 3h and Figure S11). The device also exhibited consistent responses even under > 100 cycles of applied pressure (Figure 3i).

Given the high flexibility, stretchability, and sensitivity of the 3D printed tactile sensing device, we next applied the device toward monitoring human motions. This capability was evaluated by mounting the sensor directly above the radial artery to monitor the physical force of a pulse in real time (Figure 4a). The pulses were monitored under two different conditions: (i) sedentary (~ 60 beats min^{-1}) and (ii) post-exercise (~ 120 beats min^{-1}). As shown in Figures 4b–c, the sedentary pulse showed average current changes of 80%, and the post-exercise beat showed an average current change of 118%, with higher frequencies. Therefore, pulse rates can be differentiated by discernible magnitudes and frequencies. The durability of the tactile sensor was demonstrated by wearing it for three consecutive days and repeating the pulse tests (Figure S12). The sensing behavior remained unchanged throughout the three days of testing. For further applications, the tactile sensor was mounted on a fingertip (Figure 4d) and attached to the joint of the index finger (Figure 4e) for monitoring large-scale movements. It was observed that the response signal exhibited outstanding stability and reproducibility upon both finger pressings on a keyboard and bending-release motions. These results provide a proof of concept of the abilities of the printed tactile sensor to monitor various degrees of human motions.

Next, the 3D sensor was conformally printed on a fingertip using inverse engineering technologies (Figure 4f, and Movie S4), which were realized via a combination of 3D scanning, printing, and modeling analysis. A hand model was first scanned using a 3D structured light scanning (SLS) technique (Figure S13a–b). The SLS imaging technique has been used previously to obtain the geometrical information of complex freeform objects including body parts and teeth.^[23] A 3D model with the topological information of the hand model was generated in point cloud format as shown in Figure S13c. Then, the point cloud format was transformed to 3D polygon meshes (Figure S13d–e), which were required to redesign the tactile sensor so that it was conformal to the surface of the fingertip (Figure S13f). Finally, the tactile sensor was directly printed on the fingertip by using our custom-designed multi-material 3D printing process (Figure S13g–i).

The sensing behavior of the printed tactile sensor on the fingertip upon a pressing by a human finger was then explored (Figure 4g and Movie S5). The amplitude of the output signal varied with the pressing forces applied with a time interval of 5 sec. Upon applying a weak finger press (~ 100 kPa), the sensor responded with a current change of 5000%. When a strong press was applied (~ 400 kPa), the signal intensity increased to 8000%. These distinguishable response signals also demonstrated the capability of the device for detecting

and differentiating human movements. This approach thus provides a unified and customizable manufacturing scheme for next-generation directly printed wearable devices with diverse integrated functionalities.

For electronic skin applications, it is desirable to scale up the sensor to an array configuration of several pixels to collect spatially resolved pressure information. For the purposes of this demonstration, we fabricated a 5×5 pixel array with a total area of $1 \times 1 \text{ cm}^2$ as shown in Figure 4h and Figure S14. The individual pixel had an area of $3 \times 3 \text{ mm}^2$, a height of 1.2 mm, and a common bottom electrode. To test the device, a triangular glass object (0.096 g) with a weight of 50 g was placed on top of the array, and it was observed that the magnitude of the output signal depended on the specific location of the object on the array. The color contrast mapped local pressure distributions, consistent with the shape of the triangle (Figure 4i). Importantly, our technique could be capable of providing large area and facile processing for 3D sensors or wearable biomedical devices.

In summary, we have developed a series of novel inks, which can be cured at room temperature with tunable printability, high flexibility, electrical conductivity, and sensitivity. The inks were used to manufacture 3D tactile sensors under mild conditions on a freeform surface using multi-material 3D printing and inverse engineering techniques. The printed flexible, stretchable, and sensitive sensors were found proven to be capable of detecting and differentiating human movements, including radial pulse, and finger pressing and bending. This work represents a proof-of-concept illustration that image-coupled 3D multi-material printing approaches can facilitate customized wearable devices in previously inaccessible ways. Development of a custom-built multi-functional 3D printing process, combined with functional inks is at the core of this approach and determines the features of the final devices. 3D scanning and reverse engineering allowed us to design the specific geometry to fit the curved surface. Mechanical and computational tools enabled us to design, analyze, and optimize the integrity of the devices. A one-pot multi-material 3D printing process provided us the ability to integrate various functional inks into a 3D sensor with a conformal design and high performance. This combination of complex geometries and sophisticated functions offers a proof-of-concept approach for the next generation of wearable devices. Future studies will focus on several directions, including: (i) further optimization of the inks, including incorporation of semiconducting materials and devices; (ii) development of other critical devices such as temperature sensors to monitor tissue; and (iii) development of new 3D printing platforms with closed-loop feedback control for real-time printing of objects on arbitrary and moving substrates. Overall, we expect that our methodologies will open new routes to fabricating various sensors with the potential for advancing prosthetic skins, bionic organs, and human-machine interfaces.

Experimental Section

Materials System

The silver nanoparticles and nanowires were synthesized by modifying previously published methods.^[53–55] The silver flakes and submicron particles were purchased from Sigma-Aldrich. The silicone ink was prepared from Dragon Skin ® 10 Slow Cure Part A and B, Thi-Vex Silicone Thickener and Slo-Jo ® Platinum Silicone Cure Retarder, all from

Smooth-On, Inc. Ag/silicone inks (68, 75 and 80 wt%) were prepared as follows: Dragon Skin ® 10 Part A and B were mixed at 1:1 ratio, followed by addition of Slo-Jo ® Cure Retarder (0.5 wt%) and solvent DCM (10 wt%). Then, proper amount of silver particles (0.5 – 1.0 µm) were mixed with the viscous silicone mixture. The sacrificial ink was prepared by mixing 40 wt% Pluronic F127 (Sigma-Aldrich) with ultra-filtrated water to print the supporting layer, which was removed by immersing the device in water for several hours. All the inks were mixed using a planetary centrifugal mixer (ARE-310, Thinky) and centrifuged to remove any air bubbles before printing at room temperature.

Multi-material 3D Printing

Printing was carried out using a custom-built 3D printer (AGS 1000, Aerotech) with two independent *z-axis*, which translate the printing head in the prescribed pattern. Four dispensing apparatuses (HP7x, EFD) were used to deposit the four inks through micro-nozzles with 100 µm inner diameter (5132-0.25-B, EFD). Each *z-axis* controlled two dispensing apparatuses with different heights, which ensured that the first printing tip would not touch the base when the other was printing. All printing paths were controlled using G-code commands, which were generated by the software, Slic3R from 3D models. The 3D tactile sensor model was designed using SolidWorks software. The single sensor was printed within 3 min and cured at room temperature overnight prior to characterization. The printed bottom and top electrode ends of the single sensor were bonded with a thin, flexible cable for external tests. The 5 × 5 pixel array had a common printed bottom electrode and 25 separated top electrodes for external cable connections.

Reverse Engineering of Hand Model

The individual scans were obtained using the DAVID Vision system (SLS-1, David) for scanning the hand model with 30° intervals. Then the images were aligned and assembled to a water-tight 3D model of the scanned hand model. A reverse engineering software (Geomagic) was used to transform the 3D model file from point cloud data into 3D polygon meshes, which were edited by SolidWorks for the conformal device design. Finally, the Slic3r software was used to slice the 3D model and generate the G-code for 3D printing process.

Sensing Behavior Testing

The compression force was controlled by a programmable robot (F5200N, Fisnar) with a custom-build positioner. The specimen was placed on an electronic balance (S622, H&C Weighing Systems), which can measure and transfer the real-time force value to a computer. Then the specimen was pressed by the positioner with cyclic pressures. The current differences for the sensors were acquired with a source meter unit (Model 2450, Keithley) and transmitted to a computer via a serial port. The tactile sensor mounted on the human hand was covered by a stretchable tape (VHB, 3M) for a steady connection. The sensing signals were also recorded by the source meter. An IRB review at the University of Minnesota determined that this experiment does not meet the federal definition of human subjects research and therefore an IRB review was not required.

Mechanical Testing

The mechanical properties of each ink and the printed device were studied using a dynamic mechanical analyzer (RSA-G2, TA Instruments). The samples for the tensile test were printed using various inks in the form of filaments (diameter 1 mm, length 10 mm). The specimens of each ink for the compression test were printed in the form of cylinders (diameter 5 mm, height 5 mm).

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Figure 1.

Tactile sensor design principle and 3D printing procedure. (a) Schematic of the tactile sensor consisting of a base layer, top and bottom electrodes, an isolating layer, a sensor layer, and a supporting layer. (b) Side and (c) top view of the tactile sensor. (d) 3D printing process of the sensor on a glass substrate in eight sequential steps. In step I, a $4 \times 4 \text{ mm}^2$ silicone base layer is printed. In step II, a $3 \times 3 \text{ mm}^2$ bottom electrode layer is printed using the 75 wt% Ag/silicone ink. In step III, a 1-mm-tall, 150- μm -thick cylinder wall with a radius of 350 μm is printed using the 68 wt% Ag/silicone ink as the sensor layer. In step IV, a $3 \times 3 \text{ mm}^2$ isolating layer is printed using the silicone ink. In step V, a $3 \times 3 \text{ mm}^2$ supporting layer with a thickness of 0.8 mm is printed using the 40 wt% Pluronic ink. In step VI, a $2 \times 2 \text{ mm}^2$ top electrode layer is printed using the 75 wt% Ag/silicone ink. In step VII, the supporting layer is removed by immersing the sensor in water for three hours. Finally, in step VIII, the sensor is dried for completion.

Figure 2.

Mechanical characterization and sensing behavior of various inks. SEM images show the Ag particle distribution in the inks with (a) 68 wt% and (b) 75 wt% Ag loadings. All scale bars are 5 μm. (c) Tensile and (d) compressive strength measurements on the cured inks with different Ag loading contents. (e) Resistance as a function of applied pressure for three inks. The specimens for the 68% Ag ink were cylinders (diameter 1 mm, height 1 mm), and for the 70% and 80% Ag inks were serpentine filaments (length 15 mm, diameter 0.2 mm). (f) Relative current change of 68 wt% Ag/silicone ink upon three different applied cyclic pressures. The red lines show the signals for 60 kPa applied pressure. The yellow lines are the signals for 120 kPa loading. The blue lines are the signals for 250 kPa loading.

Figure 3.

Sensing behavior of the 3D printed tactile sensor. (a) Top view SEM image of the printed sensor layer. (b) Side and (c) top view SEM images of the 3D printed tactile sensor. All scale bars are 200 μm. (d) Current-voltage characteristics of a tactile sensor under different applied pressures. (e) Plots showing frequency responses to a dynamic pressure of 200 kPa at an input frequency of 0.125 Hz. (f) Current change for various frequencies of 200 kPa applied pressure. (g) Mean compressive gauge factor for different ink materials and the tactile sensor device. (h) Current change of the tactile sensor upon applying a constant strain with an initial pressure of 500 kPa. (i) Current change of the device when subjected to 100 pressing cycles with a pressure of 100 kPa at a frequency of 0.25 Hz.

Figure 4.

Mechanical sensing applications of the 3D printed stretchable tactile sensor. (a) Photograph showing the tactile sensor mounted directly above the radial artery. Measurement signals of radial pulse under (b) sedentary and (c) post-exercise states (running upstairs for 5 min). Plots showing the current change signals in response to dynamic loading and unloading cycles in (d) pressing and (e) bending. (f) Optical image showing the conformally printed 3D tactile sensor on a fingertip. Scale bar = 4 mm. (g) Current change signal of the tactile sensor printed on a fingertip upon pressing by a human finger. (h) Top view of a triangular glass object placed on the surface of the 5×5 pixel tactile sensor. Scale bar = 2 mm. (i) Signal mapping of the pressure distribution for a triangular object (0.096 g) with a 50 g weight.