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Stretchable electronics based on Ag-PDMS composites

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Patterned structures of flexible, stretchable, electrically conductive materials on soft substrates could lead to novel electronic devices with unique mechanical properties allowing them to bend, fold, stretch or conform to their environment. For the last decade, research on improving the stretchability of circuits on elastomeric substrates has made significant progresses but designing printed circuit assemblies on elastomers remains challenging. Here we present a simple, cost-effective, cleanroom-free process to produce large scale soft electronic hardware where standard surface-mounted electrical components were directly bonded onto all-elastomeric printed circuit boards, or soft PCBs. Ag-PDMS tracks were stencil printed onto a PDMS substrate and soft PCBs were made by bonding the top and bottom layers together and filling punched holes with Ag-PDMS to create vias. Silver epoxy was used to bond commercial electrical components and no mechanical failure was observed after hundreds of stretching cycles. We also demonstrate the fabrication of a stretchable clock generator.

Stretchable electronics enables new applications such as stretchable displays[1], electronic skins[2], flexible sensors for personalized healthcare[3] and conformal electrode arrays to interface the heart and brain[4] that are not possible with rigid electronics. To achieve this, standard silicon or polymeric substrate materials were replaced by elastomers and one of the key technical challenges was to develop stretchable electronic materials that offer the electrical conductivity of metals while enduring large repeated strains. Several promising approaches have been developed to improve stretchability. One was to play with the geometry of inorganic conductors to compensate the mechanical properties mismatch with the elastomeric substrate either by using thin gold films[5], wavy silicon nanoribbons[6] or by patterning spring-shaped metal traces[7,8]. Another approach consisted in shaping the substrate e.g. making it porous or engineering microfluidic channels and coating it with a metal layer[9] or filling it with liquid metal alloys like eutectic GaIn[10,11]. A third approach was to produce stretchable conductive composites by dispersing[12], casting[13], reducing[14] or implanting[15] nano or micro conductive particles in poly(dimethylsiloxane) (PDMS) or polyurethane (PU). Although these methods are ingenious and useful for producing stretchable interconnects, they do not offer all the possibilities that printed circuit boards do. Large-scale production of solderable stretchable electronics using standard design and routing tools that can be interfaced with rigid electronics remains a challenge but would make this technology even more attractive.

In this paper we present a simple, low-cost and large scale process to produce all-elastomeric solderable stretchable printed circuit boards that we refer to as soft PCBs and we show a few stretchable circuits using commercial electrical components. The stretchable conductor used in this method, which is a mixture of Ag and PDMS, can be stretched at high strains while maintaining a high conductivity[12]. Here, the electrical and mechanical properties of the composite were investigated to determine the best mixture for stretchable circuit applications. The conductivity, contact resistance and response to various applied strains as well as elastic modulus, maximum tensile strain and Poisson ratio of samples with different Ag volume contents were measured. We demonstrate that this composite can be stencil printed or screen printed with a resolution of 150 μm for large scale fabrication of large stretchable circuit boards on PDMS substrates. Double sided soft PCBs with vias were successfully produced by bonding the bottom and top layers and filling punched holes with the same composite to create vias. The conductivity of the printed composite surprisingly increased with the reduced wire dimensions. We believe this is due to the fact that particles are brought closer to each other during the printing of narrow lines. The tracks of a double sided soft PCB had a typical resistance of 2 Ohms/cm.

Direct bonding of commercial electrical components on a stretchable substrate is challenging and usually requires the substrate to be stiffened[16–18] or the component to be encapsulated[19]. Recently electroplated Ni on a conductive porous PDMS was used as a metal anchor for affixing LEDs[9]. However this process involved several metal deposition and electroplating steps and required meander shaped tracks for stretchability. Here we demonstrated the use of Ag-epoxy to bond commercial electrical components on straight tracks for the construction of

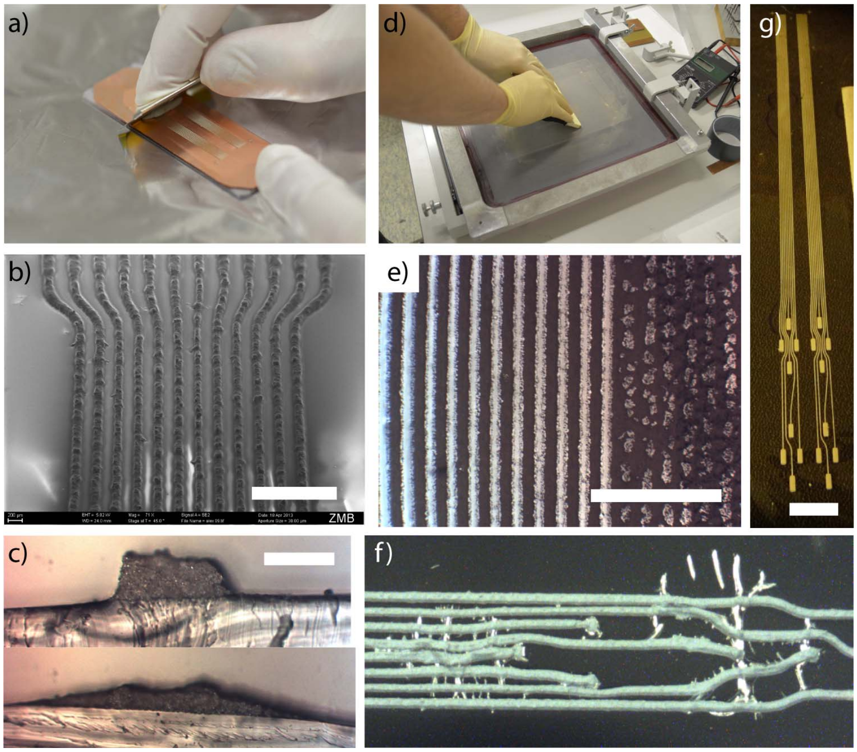

Figure 1 | **Fabrication of soft PCBs using stencil printing and screen printing.** (a) Picture showing the stencil printing of Ag-PDMS composites. (b) SEM picture of the stencil printed lines with highest achieved resolution. (c) Microscopic images showing the cross sections of stencil printed Ag-PDMS with 25vol% (top) and 13vol% (bottom). (d) Picture showing the setup for screen printing on 8″ wafers. (e) Microscopic image of finest screen printed lines (black) and spacing (white). (f) Microscopic image showing typical defects like short circuits or delamination of the PDMS layer from the glass substrate. (g) Picture of very long screen printed tracks. Scale bars are 0.1 mm in (c), 1 mm in (b), (e) and 10 mm in (g).

electronic circuits for the first time as it is actually done with PCBs. We also show the compatibility of our soft PCBs with ZIF connectors to interface soft and hard circuits together.

Results and discussion

Fabrication of soft PCBs using stencil printing and screen printing. *Stencil printing*. Large-area Ag-PDMS structures were fabricated using standard industrial printing techniques: stencil printing and screen printing (see Methods). Custom-made copper stencils were used as shadow masks for patterning structures of Ag-PDMS as shown in Fig. 1a. The adhesion between the PDMS substrate and the stencil was affected by the roughness of the copper surface. Stencils with smooth surface (no treatment) adhered strongly to the substrate and did not move during printing whereas thinner stencils produced by wet etching from both sides poorly adhered to the PDMS due to their rougher surface. Hence, thinner stencils were produced by wet etching one side of the copper stencil while protecting the other side with tape. 150 μm wide lines with 100 μm spacing were achieved using 100 μm thick stencils as shown in Fig. 1b. Such stencils were robust enough to be reused several times without getting deformed contrary to thinner stencils that were more fragile. Hexane was used as a cleaner between consecutive printings. Printing could be repeated several times using the same stencil without losing quality. Mishandled stencils tended to buckle and did not lay perfectly flat during printing which led to defects and unwanted short circuits between lines. Figure 1c shows the cross sections of lines printed using Ag-PDMS with different viscosity. High resolution patterns could not be achieved using Ag-PDMS with filler content below 22vol% since the material was flowing after the stencil was removed due to its lower viscosity. Prolonged use of Ag-paste during repeated printings accelerated the cross linking and aging of the composite thus increasing excessively the viscosity of the paste and introducing short circuits between lines. However, Ag-PDMS pastes could be stored at −24°C over a year without dramatic loss of performance. Figure 1f shows typical short circuits between lines resulting from bad printing. Such defects could be manually removed before the composite was cured or after curing by cutting away the undesired parts, as it is done with standard PCBs, and filling the cavities with uncured PDMS. Closed lines could not be printed using this method because there was no frame to hold the stencil together.

Screen printing. This was not anymore a problem when using screen printing since the screens are composed of a mesh on which a thick resin is applied. The screen was fixed on a frame and its vertical position could be adjusted with screws. The distance between the screen and the PDMS was set to 4 mm to prevent the resin to stick to the PDMS during printing. Hence a metallic mesh was preferred to a plastic mesh because of its superior rigidity to avoid buckling after such a large deformation. Large patterns could successfully be printed on 8″ wafers covered with PDMS as shown in Fig. 1d. Line widths of 70 μm and spacing of 50 μm were achieved as shown in Fig. 1e. Lines were 40 μm thick, which corresponds to the thickness of the resin on the screen. Smaller features could not be fully transferred and looked like dashed lines. Cleaning the mesh before each printing was necessary to remove residues clogging the mesh apertures. Figure 1g shows large soft PCBs produced using screen printing. The PCBs were soft enough to conform to various surfaces.

Patterning conductive PDMS on elastomeric substrates have been investigated in quite a few ways by microcontact printing[20], trench filling[21] or photocrosslinking conductive PDMS[22]. High resolution patterns of highly viscous pastes can be achieved using trench filling or photopatternable conductive PDMS but are challenging using microcontact printing. stencil printing and screen printing are commonly used in the electronics manufacturing process to print electronic circuits. Although large stencil printed patterns of Ag-PDMS were already reported by others[23], we investigated here the feasibility of printing stretchable circuits of dimensions comparable to standard electronics. We also showed that screen printing was compatible with this material and that clogged Ag-PDMS can be removed to allow multiple printing.

Electro-mechanical properties of Ag-PDMS composite. Percolation in composite materials depends on the filler material, aspect ratio, quality of dispersion and matrix material and influences both electrical and mechanical properties. Conductivity and contact resistance were measured using Kelvin sensing on stencil printed Ag-PDMS stripes (40 mm × 5 mm × 0.1 mm) with different filler volume fractions ranging from 12% up to 25%. Cured samples were peeled off, placed on a glass slide and four probes were pressed against the sample using magnets. The volume content of Ag particles determines the size of percolation networks in the PDMS matrix and the conductivity of the composite. Electrical conductivity of 1 S/cm was measured in samples with 13vol% and then rapidly increased to 100 S/cm when adding small amounts of silver and finally reached 600 S/cm after loading 25vol% of silver in the PDMS matrix. The contact resistance followed the inverse behavior as shown in Fig. 2a. The data were best fitted with the following percolation model (red line) $\sigma_c = \sigma_0(c - c_t)^t$ where σ_0 was 18168 S/cm, c_t was 12.6% and t was 1.68. The changes in resistance R/R_0 under quasi-static uniaxial strain were measured similarly in a tensile test machine at the rate of 1%/min. Rapid increase in resistance occurred in samples with low silver content when samples with higher concentrations were stretched above 100% as shown in Fig. 2b. Samples with 23vol%, 24vol% and 25vol% were still conductive before rupture. Large dog-bone samples of Ag-PDMS 13vol%, 16vol%, 19vol%, 22vol%, 25vol% and pure PDMS were molded and mounted in the tensile test machine to measure its mechanical properties. Figure 2c shows stress strain curves for a strain rate of 0.1 mm/s. The elastic moduli were defined as the slope for the first 1% and are plotted against the silver content in Figure 2d. The large error bar for 25vol% can be explained by the outlying behavior of one of the three samples that showed higher stiffness and early fracture. The max strains at break are shown in Figure 2e. It appears that samples loaded with silver particles exhibited a higher strain at break that increased with the silver volume content and was maximal for 19%. Adding more silver seemed to fragilize the material and decrease the maximal elongation at rupture. Excessive amount of silver also increased the viscosity, thus making the printing difficult and the samples more brittle. To investigate the changes in Poisson ratio with silver content, Ag-PDMS dog-bone samples were fixed on a custom-made manual stretcher placed under a measurescope. Changes in width were measured while stretching uniaxially. The Poisson ratio was lower for composites with higher filling contents and decreased with applied strains as shown in Fig. 2f. Elastomers like PDMS are generally considered to be incompressible with a value of Poisson ratio of 0.5. Increasing the amount of fillers decreased the Poisson ratio that was shown to be strain dependent probably due to dewetting and vacuole formation[24] due to weak bonding between the silver particles and PDMS. Under tensile strain the volume of the composite increased, which decreased the volume content of Ag towards the percolation threshold, which explains the rapid increase in resistance. The influence of sample sizes on conductivity and stretchability was also investigated. Narrower tracks (2 mm, 1 mm, 0.6 mm, 0.3 mm, 0.25 mm, 0.2 mm and 0.15 mm) were stencil printed on the same substrate, cured, peeled off and placed on a glass slide. Figure 3a and 3b show resp. the max current densities and sheet resistances as a function of the line width. The sheet resistances unexpectedly decreased when reducing the track width, which is in contradiction with percolation theory. The electrical resistance of a composite conductor should increase when its dimensions become comparable to the size of the filler particles because of the lower probability of finding conductive pathways. The increase in maximum current density is consistent with the decrease in sheet resistance. The mean conductivity for each line width was plotted against the volume fraction in Fig. 3c (without error bars for clarity) and the data were fitted using the previously introduced percolation model with the same values of c_t ant t. The fitted apparent conductivity significantly increased when the track width was decreased below 0.5 mm as shown in Fig 3d. These results suggested that the conductivity of the material was influenced by the size of the printed patterns. There are at least two possible explanations to this effect. The particles may come in closer contact with each other thus lowering the resistance at the interface because of higher compressive forces when printed through a narrower mask or an increase in particles concentration due to the lateral flow of PDMS after printing. The samples of the first batch of Ag-PDMS 25vol% with different lines widths were stretched to 50% at a speed of 1 mm/s and the change in resistance was plotted in Fig. 3e. The narrower lines showed less increase in resistance, which is consistent with their previously described superior performances. Standard PCBs often have copper traces of 200 µm in width. These results suggest that printing stretchable conductive circuit boards with track widths similar to that of rigid PCBs is feasible. For the production of double-sided soft PCBs, the top and bottom layers were bonded together and vias were created by punching holes through the board and filling them with the same Ag-PDMS paste as shown in Fig. 3f.

Another method for creating through silicone vias reported elsewhere[23] consisted in bonding patterned layers of Ag-PDMS to Ag-PDMS vias printed onto a PMMA substrate and filling the gap with uncured PDMS using a syringe. Here, we aimed at providing a method similar to what is found in the industry where drilled holes are filled with a conductive paste. This approach was also successfully used with polyurethane composites for flexible electronic applications[25].

Bonding components and interfacing soft electronics with hard electronics. Interconnecting electrical circuits can be done in a reversible manner using mechanical clamping or in an irreversible way using solder bonding. Zero Insertion Force (ZIF) connectors were used to interconnect soft PCBs to rigid standard PCBs. A stretchable ribbon cable with 8 leads was produced using stencil printing and was clamped between two ZIF connectors as shown in Fig. 4a. The ZIF connectors provided good electrical contacts even when the ribbon cable underwent large strains as high as 40% (see Supplementary Movie 1) but mechanically damaged the printed tracks after repeated manual stretching cycles due shear stress exerted by the metallic contacts on the soft Ag-PDMS leads. To solve this problem, the local stress can be delocalized by reinforcing the terminals with a sheet of polyimide or additional clamping on the PDMS. Novel designs of ZIF connectors including a second clamping system could significantly improve the reliability of the connector. Another way of interconnecting was to use Ag epoxy to bond the soft ribbon cable onto a rigid double-sided PCB. Figure 4b shows the interconnection between a stretchable ribbon cable with 12 conductors and a miniature custom-made connector. This solution allowed for smaller contact area and miniaturized interconnections but the bond is permanent. The Ag epoxy bonding technique was also used to mount SMD components onto the soft PCB. Chip resistors of various sizes were bonded between two tracks with different widths. Figure 4c shows 0406, 0603 and 0805 chip resistors after bonding. The samples were stretched to 20% over a thousand of cycles several cycles at 1 mm/s and the bond did not fail mechanically. When chip resistors were manually removed from the circuit by pulling on them some Ag-PDMS came off with the components suggesting good adhesion. High yield of the bonding

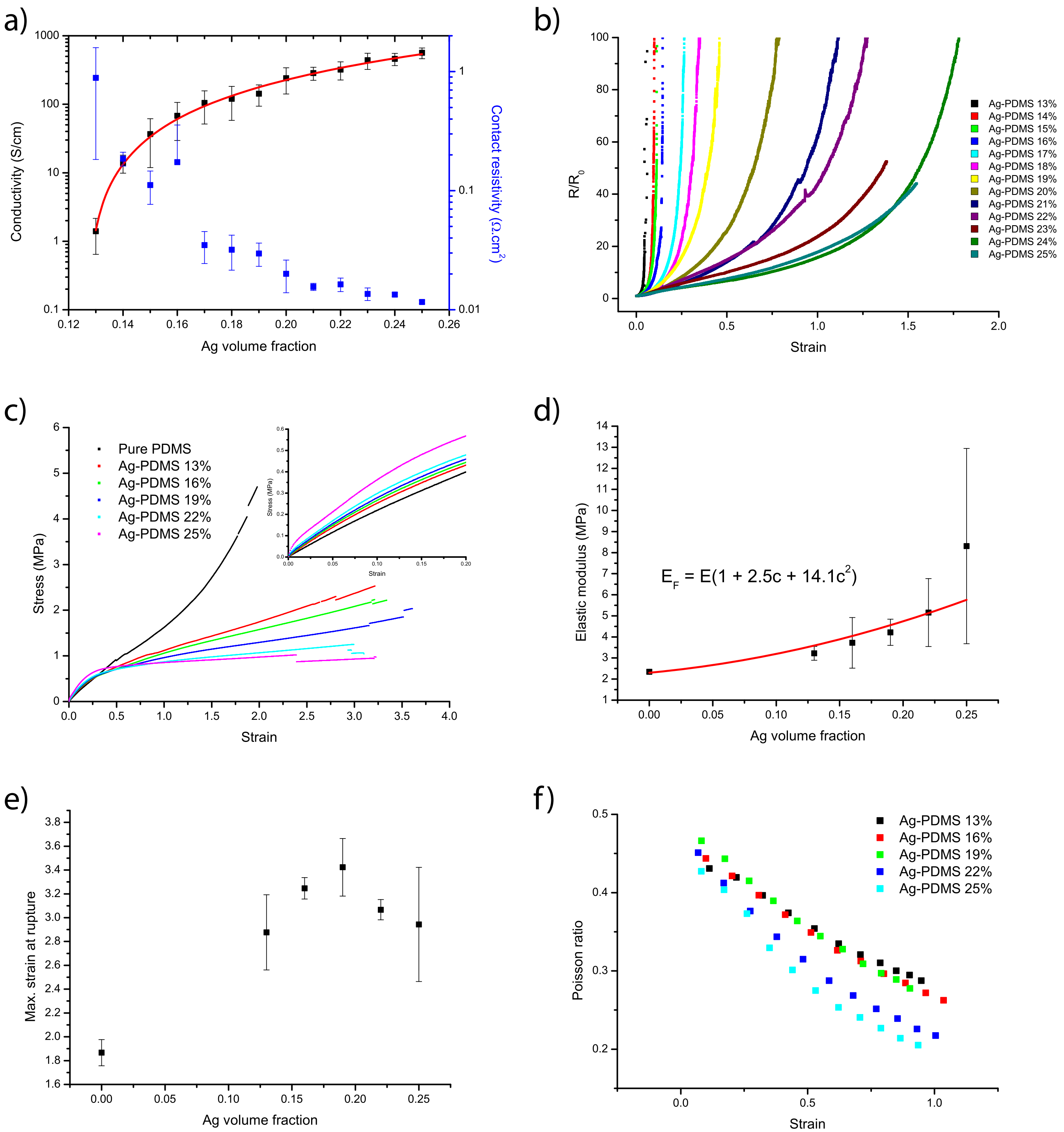

Figure 2 | Electro-mechanical properties of different Ag-PDMS composites. (a) Electrical conductivity and contact resistivity of Ag-PDMS composite as a function of silver volume fraction (n=3). Fitted parameters of the percolation model (red line) are $\sigma_0 = 18168$ S/cm, $c_t = 12.6\%$ and $t = 1.68$; adj. R-squared is 99.2%. (b) Changes in resistance during quasi-static stretching tests. (c) Stress-strain curves at low strain rate for pure PDMS (black), Ag-PDMS 13vol% (red), 16vol% (green), 19vol% (dark blue), 22vol% (light blue) and 25vol% (pink) (n=3, error bars removed for clarity). Elastic moduli were derived from the slope at 1% strain (inset) and (d) plotted vs silver volume content. Data are well fitted with the Guth-Smallwood equation. *E* is the elastic modulus of pure PDMS and *c* is the silver volume fraction. (e) Maximal strain at rupture as a function of filler content (n=3) and (f) measured Poisson ratios for different silver contents.

was demonstrated with 6 × 7 arrays of LEDs = 84 contacts (see Fig. 4d). A soft astable circuit generating a 2 kHz clock was produced using the described method and is visible in Fig. 4e. Finally, a stretchable clock generator assembled on a double-sided PCB including vias with LEDs flashing every second and a ZIF connector is showed in Fig. 4f. The frequency of the clock determined by the values of the resistor and capacitor remained stable during manual bending and stretching (see Supplementary Movies 2 and 3).

Conclusions

The proposed method integrates important standard PCB design features like straight traces, vias, solderability, connectivity with

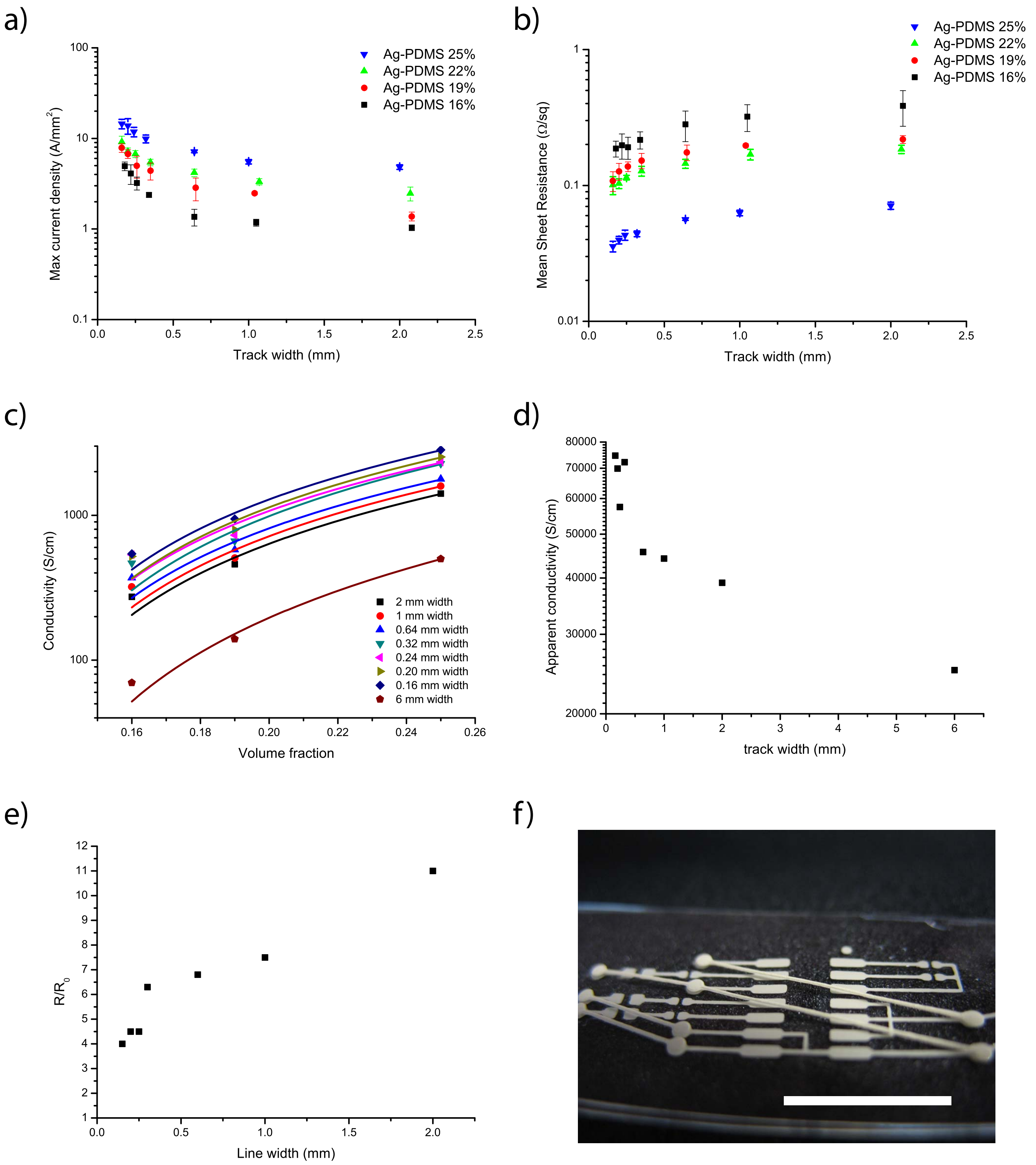

Figure 3 | Stencil printing of narrow lines for the fabrication of soft PCBs. (a) Maximum current densities and (b) sheet resistances for Ag-PDMS tracks with different widths and filler contents. (c) Mean values of measured conductivities for different tracks as a function of silver volume fraction and fitted using the percolation model with the apparent conductivity as only variable. (d) The fitted apparent conductivity as a function of track width. (e) Change in resistance after 50% strain 1 mm/s for Ag-PDMS 25vol% tracks with different widths. R is the resistance after releasing the strain and R_0 is the resistance before applying the strain. (f) Picture of a double-sided PCB with vias. The large footprint in the center is for the soldering of a SOIC 14 pins packaged IC. Scale bar is 10 mm.

standard hardware and can be used to design soft and stretchable PCBs the same way rigid or flexible PCBs are designed. Stencil or screen printing Ag-PDMS is a simple solution for large scale production of PCBs at low costs and the method of bonding them together and making vias for double sided PCBs is convenient and similar to current industrial processes. Double-sided soft PCBs had in average a low ohmic resistance of 2 Ohm/cm. Commercial electrical components can be bonded onto Ag-PDMS tracks using a

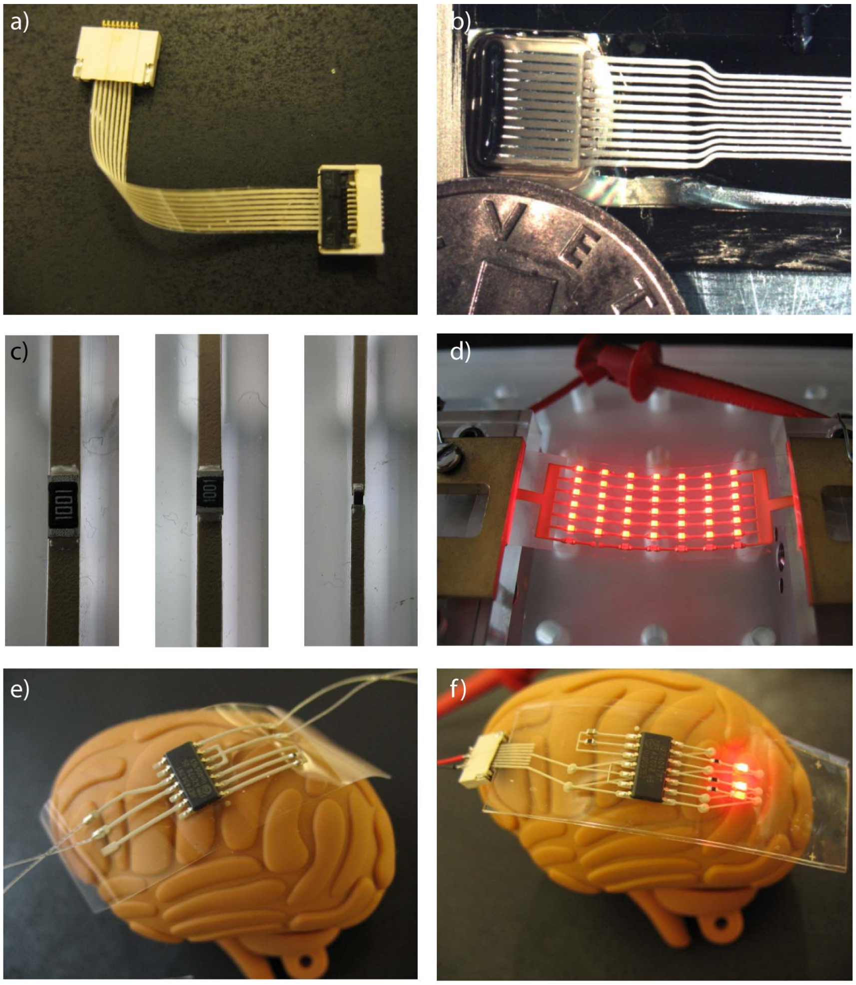

Figure 4 | Fabrication of soft electrical circuits on PDMS integrating commercially available electrical components. (a) Picture of a ribbon cable with 8 conductors clamped on both ends by a commercial ZIF connector. (b) Picture of a custom-made miniature connector with 12 contacts. (c) Pictures of chip resistors bonded on conductive tracks (from left to right: 0805, 0603 and 0402 packages). (d) Picture of a large array of SMD LEDs bonded on a soft PCB. (e) Picture of a 2 kHz clock generator produced on a 0.2 mm thick soft single-sided PCB that conforms to a plastic brain. (f) Picture of a 1 Hz clock generetor with LEDs to display the output levels and produced on a 0.7 mm thick double-sided soft PCB (that shows less conformability on the plastic brain than the single-sided PCB) with vias and connected to a power supply with a ZIF connector.

standard Ag-epoxy to fabricate soft circuits. These circuits can be interfaced to rigid electronics using commercial ZIF connectors. An astable circuit including all these features was made to demonstrate the presented method. This technique also enables the fabrication of thin, soft and stretchable conductive leads for multielectrode arrays with mechanical properties closer to tissue than state-of-the-art polyimide-based neural interfaces. Such silicone-based implants can be used to stimulate or record from the brain or the spinal cord without damaging the delicate neural tissue even when implanted in the subdural region. This technology paves the way for a new generation of neuroprosthetic devices.

Methods

Fabrication of the Ag-PDMS composites. To fabricate the Ag-PDMS composites, the PDMS prepolymer (Sylgard 184) and silver powder, 2–3.5 micron, 99.9+% (Sigma Aldrich) were first dispersed by hand using a spatula and then mixed in a planetary mixer (Thinky ARE-250) for 3 minutes at 2000 rpm and degassed for 1 minute at 2200 rpm. Next, PDMS curing agent was added (respecting a 10 : 1 ratio between the prepolymer and curing agent) and incorporated into the mixture by stirring manually before mixing again for 1.5 minute at 2000 rpm and degassing for 30 seconds at 2200 rpm. The viscous paste that was obtained was then stored in a freezer at -24°C until usage. Prior to usage, the paste was manually stirred with a spatula and mixed for 1 minute at 2000 rpm and degassed for 30 seconds at 2200 rpm.

Stencil printing and screen printing. Copper stencils were produced on site by wet etching of copper foils (Goodfellow, England). To print Ag-PDMS structures on a PDMS substrate, the Ag-paste was forced into the stencil using a tape-covered razor blade as a squeegee hold to a 45° angle with the stencil. The applied pressure was just enough to remove the excess paste on the stencil. The stencil was then gently peeled off. Hexane was used to clean the stencil. Ag-PDMS was screen printed in a similar manner using screens purchased by Mantel Digital AG. A metallic screen with 400 mesh and 40 microns thick resist was hold 4 mm far from the PDMS substrate. A hard squeegee was used to spread and press the paste into the mesh. The screen was cleaned using a cleaner solution (Mantel Digital AG, Wädenswil, Swizerland). All printed samples were placed in an oven at 80°C for 4 hours and let cool down to room temperature.

Mechanical characterization. A measurescope was used to measure the widths of printed narrow lines. A microscope was used to measure the thickness of printed lines by focusing successively the upper and lower surfaces. Stress strain curves were obtained using a tensile test machine (DO-FB0.5TS, Zwick Roell, Germany).

Electrical characterization. Electrical resistances were measured using four-terminal sensing. A stabilized voltage source was used to deliver current to the probe. The current was measured over a sense resistor with value chosen in the same order of magnitude than the resistance of the probe. The resistances of the probe and of the contacts were derived from the measured voltage drop over the contacts and the probe.

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Author contributions

A.L. designed and performed most of the experiments, analyzed the data, prepared the figures and wrote the manuscript. S.E. was involved in the tensile tests measurements and results analysis. H.J. was involved in setting up the screen printing technique and tested the technique. A.L. and J.V. discussed the results and commented on the manuscript.

Additional information

Supplementary information accompanies this paper at http://www.nature.com/scientificreports

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