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Soft Electronic Platforms Combining Elastomeric Stretchability and Biodegradability

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Supporting Information

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**S1. Detailed synthesis**

**S1.1. Synthesis of Poly(glycerol sebacate)**

The poly(glycerol sebacate) (PGS) polymer was synthesized by a polycondensation reaction of glycerol (Alfa Aesar, ThermoFisher, Kandel, Germany) and sebacic acid (Merck KGaA, Darmstadt, Germany). The reagents were added in equimolar amounts into a flask and stirred at 135°C under an argon atmosphere for 1 h 20 min, until the reagents formed a homogenous liquid mixture of medium viscosity. The colorless mixture was stirred for additional 2 h at 120°C under argon atmosphere. Then a vacuum of ~2 mbar was applied for 72 h at 120°C under stirring. Over this time, the mixture changed to a yellow color and the viscosity increased. The polymer was used without purification.

The variation results from taking samples from different regions in the flask. Despite thoroughly stirring the melt, the top part of the melt is likely to contain shorter chains, as they melt at lower temperatures. The matrix-assisted laser-desorption-ionization time-of-flight mass spectrometry (MALDI-TOF, Bruker Autoflex Speed) data peaks at a lower mass to charge ratio of 0.2 kg/mol, which may indicate that there are 2 to 3 charges per fragment ([Figure S1 a](#)). Alternatively, assuming 1 charge per fragment, the lower peak position may indicate a polymer fragmentation by the laser during the ionization process. The MALDI-TOF spectra of PGS batch1 and 2 match, however, highlighting a reproducible PGS synthesis procedure.
S1.2. Synthesis of Poly(glycerol sebacate) acrylate

**Figure S1.** Molecular weight distribution of (a) PGS and (b) PGSA as measured by MALDI-TOF (peaks) and GPC (curves).

PGS serves as a pre-polymer for PGSA, which was synthesized by an esterification of PGS with acryloyl chloride. The amount of acryloyl chloride determines the degree of acrylation (DA) in the PGSA. In two different batches of PGSA, the nominal DA was set to 16% (PGSA-19) and 24% (PGSA-28). The required amount of acryloyl chloride was calculated from the desired DA and selected amount of PGS pre-polymer via an empirical equation:

\[
DA = 0.655 \times \frac{\text{mol (acryloyl chloride)}}{\text{mol (PGS)}} + 0.0005
\]

The final DA turned out to be 19% (PGSA-19) and 28% (PGSA-28), as described in section S1.3. The arising hydrochloric acid was intercepted by triethylamine (TEA), being added in equimolar amounts to the acryloyl chloride. 4-(dimethylamino)pyridine was added as a
catalyst for the esterification and 4-methoxyphenole was added as an inhibitor for a radical polymerization of the acrylic groups.

Table S1. Number- and weight averaged molecular weight values for different PGS and PGSA batches (from GPC data in Figure S1).

| Sample Batch | $M_n$ (kDa) | $M_w$ (kDa) |
|--------------|-------------|-------------|
| PGS Batch 2 sample 1 | 1.70 | 1.84 |
| PGS Batch 2 sample 2 | 0.66 | 1.06 |
| PGS Batch 2 sample 3 | 0.42 | 0.71 |
| PGSA-19 Batch 10 (first peak "++") | 0.05 | 0.07 |
| PGSA-19 Batch 10 (main peak ) | 0.68 | 0.78 |
| PGSA-19 Batch 10 (last peak "+++") | 4.75 | 6.56 |

For the PGSA batch with a DA of 19% a flask was filled with the pre-polymer PGS (10.03 g) and heated under vacuum until the visible evaporation of water stopped. The flask was then flame dried and flushed with Argon. The PGS was completely dissolved in 700 ml anhydrous dichloromethane (DCM). It was essential to perform the acrylation with a low concentration of PGS, because prior syntheses with higher concentrations resulted in a premature crosslinking of PGSA in DCM solution in less than a day. After the addition of a solution of 4-methoxyphenole (Merck KGaA, Darmstadt, Germany, to stabilize the acryloyl chloride) in dichloromethane (180 µl, 19.4 µmol, 13.3 g/l), 4-(dimethylamino)pyridine (Merck KGaA, Darmstadt, Germany) (DMAP) (11.0 mg, 90.3 µmol) and triethylamine (1.31 ml, 9.46 mmol), the solution was cooled to 0°C under a positive pressure of argon. Under the exclusion of light, a solution of acryloyl chloride in dichloromethane (8.48 ml, 9.46 mmol, 10 vol%) was added over the period of one hour. The mixture was stirred for 24 h and allowed to reach room temperature. Afterwards a solution of 4-methoxyphenole in dichloromethane (180 µl, 19.4 µmol, 13.3 g/l) was added to stabilize the acrylic group in PGSA.

For a higher DA of 28%, the same procedure was conducted with PGS (10.43 g) in 700 ml DCM, using solutions of 4-methoxyphenole in (190 µl, 20.1 µmol, 13.3 g/l), DMAP (11.5 mg,
93.9 µmol), TEA (2.05 ml, 14.76 mmol), acryloyl chloride (13.24 ml, 14.76 mmol, 10 vol%) and 4-methoxyphenole (190 µl, 20.1 µmol, 13.3 g/l) in DCM.

Despite the high dilution during synthesis, some prematurely crosslinked polymer particles formed in the flask during the acrylation and while it was stored in the fridge. The number of particles was higher for the 28% acrylation. The particles had a size of 1-5 mm but they were obviously swelled up with the inclusion of DCM. To prevent problems during the subsequent purification, the particles were filtered prior to the purification.

S1.3. Purification of Poly(glycerol sebacate) acrylate

![Figure S2](image.png)

**Figure S2.** Crystalline impurities of TEACl in a dropcast PGSA film, (a) without purifying the synthesis product, (b) after an inadequate filtration procedure according to Ifkovitz et. al.[2]

A purification step of the polymer solution was needed to remove TEACl from the solution. Otherwise the TEACl forms needle-shaped crystals during the manufacturing of films, i.e., when dropcasting the PGSA solution in DCM (**Figure S2 a**).

Ifkovitz et al. and Nijst et al. purified PGSA by filtration after the addition of ethyl acetate (EA) and removal of DCM to precipitate the salt.[1,2] For filtration two methods were tested: vacuum filtration with filter paper or through Celite. Both methods removed the visible precipitation, but after processing of the polymer, it came clear that both methods were insufficient to remove the salt completely (**Figure S2 b**). A filtration of the concentrated solution under pressure with a PTFE or PVDF syringe filter wasn’t possible because of the high viscosity of the solution.
Another strategy was to extract the polymer from the solution, using the different solubility of the polymer and TEACl. The attempt to extract the TEACl with water directly from the solution in DCM failed and ended up in a single cloudy layer with no separation at all. Instead, dichloromethane was removed to a tenth of the volume under reduced pressure in a rotary evaporator, then the original volume of ethyl acetate was added to the solution and the remaining DCM was removed. A colorless precipitation of TEACl and prematurely crosslinked polymer formed overnight in the fridge that was separated from the EA solution by filtrating the solution. Remaining TEACl crystals were removed by filtration through a filter paper, flushing the filter with excess EA. The EA solution was washed with an equal volume saturated NaCl solution (higher density than distilled water), which was enhanced with 0.5 vol% of an aqueous 10% HCl solution to react with the remaining TEA. EA produces a clearer separation of the organic and aqueous layers than DCM. The EA layer was washed once more with fresh NaCl-HCl solution. As PGSA is dispersible in water, it was subsequently extracted 5 times with fresh EA from the aqueous phase. After all organic layers were combined, dried over sodium sulphate and filtrated, the ultra-low concentrated solution can be stored without the risk of premature crosslinking. The majority of the solvent was removed in a rotary evaporator until a slightly yellow liquid with a high viscosity (10 g original weight of PGSA in approximately 24 mL EA) remained. The final solution was stored at 7°C under the exclusion of light. Before film manufacturing, the PGSA solution in EA was concentrated once more by rotary evaporation. As a bonus, PGSA solutions in EA resulted in more homogeneous films than in DCM.

The molecular weight distributions of PGSA-19 by gel permeation chromatography (GPC) and MALDI-TOF are broadened compared to PGS, hence indicate the development of higher molecular weight chains ($M_n = 4.75$ kDa by GPC) due to premature crosslinking of PGSA (Figure S1 b, Table S1). However, the GPC main peak ($M_n = 0.68$ kDa) is still clearly visible and matches the $M_n$ of PGS well. The prominent first peak (marked with “+”) corresponds to
$M_n = 0.05$ kDa and can be attributed to clouding of the solution and possibly residual TEACl crystals.

**Figure S3.** $^1$H Nuclear magnetic resonance spectra of (a-d) PGSA-19, using two independent samples (a+c and b+d), ethyl acetate peaks are marked with *. (a-b) Single re-dissolution with CDCl$_3$. (c-d) Triple re-dissolution with CDCl$_3$. (e) NMR spectrum of PGSA-28.
Figure S4. Zoomed-in $^1$H-NMR spectra of Figure S3, with acrylate-peaks on the left side and alkyl-peaks of sebacic acid on the right side. (a-d) PGSA-19, (e) PGSA-28. The roman numerals label the peaks.

Nuclear magnetic resonance (NMR) spectra were recorded with a Bruker Avance III at 300 MHz and 27°C. The used PGSA solutions were identical with the purified solutions used for fabricating elastic substrates and hence dissolved in ethyl acetate in order to obtain the final DA after the purification procedure. A single removal of ethyl acetate and re-dissolution in deuterated chloroform (CDCl$_3$) of two independent samples of PGSA-19 resulted in spectra
with residual ethyl acetate peaks (* in Figure S3, S4). The multiplet peak integral (iv) at roughly 1.6 ppm of four H-atoms of the alkyl backbone of sebacic acid was used as a reference and fixed to be 4. With this reference, the peak integral (v) at 1.2 ppm of the central eight H-atoms of sebacic acid amounted to around 8, confirming the validity of each spectrum (Figure S3, S4). The summed peak integrals of positions (i), (ii) and (iii) at 5.9-6.5 ppm, corresponding to the three H-atoms of the acrylic group, would amount to 3 at full acrylation. For the two independent samples of PGSA-19, the summed peak integrals of 0.67 and 0.49 correspond to a DA of 22% and 16%, which were averaged to DA = 19% for PGSA-19 (Figure S3 a, b, S4 a, b). Further removal of the ethyl acetate residues by repeated dissolution in chloroform and subsequent evaporation caused premature crosslinking of the acrylate groups and reduction of measured DA to 5% for both samples (Figure S3 c, d, S4 c, d). The summation of the integrated acrylate peaks of 0.85 resulted in a DA = 28% for PGSA-28 (Figure S3 e, S4 e).

S1.4. Crosslinking mechanisms of PGSA

For crosslinking via UV-light, the radical initiator 2,2-dimethoxy-2-phenylacetophenone (DMPA) was added to the purified and concentrated PGSA solution. The concentration of PGSA in the solution could not be quantified, but it was approximately a 1:1 solution in EA. Since the exact concentration was unknown, the amount of added DMPA varied from sample to sample. To approximately 4 ml of PGSA solution, around 0.1 g of DMPA solution (10% in ethanol) was added, yielding a soft elastomer. Adding 0.4 g of DMPA solution yielded haptically harder elastomers that break at a lower elongation. The solution was stored at 7°C under the exclusion of light and ready to use for the manufacturing of elastic films.
Scheme S1. Possible termination reactions of the radical crosslinking without DMPA: (1) Radical attack of a radical acrylic group onto the PGS backbone. (2) Combination of two acrylic groups.

The photo-crosslinking mechanism in the PGSA films employs a radical polymerization of the acrylic groups. The unsaturated double bonds of the acrylic groups can undergo a self-crosslinking through autoxidation with the formation of hydroperoxide as an intermediate reactive species, resulting in a bond to the PGS backbone or another acrylic group (Scheme S1).[^3]^[4]

Scheme S2. Light-induced formation of radicals of DMPA.

DMPA was added to speed up the process of crosslinking, forming two radicals when irradiated with UV-light (Scheme S2). Those radicals will react with the acryloyl groups and start the radical polymerization by creating a radical on the side chain. As the polymer backbone can’t move freely due to the entanglement with other chains, there is a high chance that the radicals get saturated by a radical acryloyl group of a nearby polymer chain, resulting in 2 possible isomers (Scheme S3). Option 4 has a higher probability than option 3, since
secondary acryl radicals are more stable. A radical acryloyl groups may also react with a non-radical nearby acryloyl group (option 5 in Scheme S3, where the radical crosslink is locally fixed by the sterically immobile polymer backbone and remains a radical until an initiator radical terminates it. Termination by oxygen is also an option.

Scheme S3. Possible termination reactions of the radical crosslinking in the presence DMPA by a combination of: (3) two primary or (4) two secondary radical acryloyl groups, (5) a secondary radical acryloyl group and a non-radical acryloyl group that eventually gets terminated by an initiator radical. \( R' \) denotes the two initiator radicals of DMPA.
S2 Film fabrication and crosslinking

S2.1. Thin film fabrication and crosslinking of PGS

A piece of the pre-polymer PGS was flattened to a 1.3 mm thick film and simultaneously crosslinked in a nanoimprinter (NIL CNI V1.0) for 72 h at a pressure of 5.5 bar and a temperature of 120°C between two Teflon sheets. Subsequently it was cut to 25x25 mm². Merely heating a solution-cast PGS layer in an oven resulted either in strong dewetting (acrylic glass) or in strong adhesion to the substrate material (glass), in combination with non-closed films. Thicker melt-cast slabs of PGS (> 1 mm) formed bubbles during the thermal crosslinking in an oven.

S2.2. Thin film fabrication and crosslinking of PGSA

The fabrication of PGSA thin films (0.7 to 1.5 mm thickness, 19% or 28% DA) was conducted by drop casting a concentrated and highly viscous solution of PGSA in EA, with added DMPA, onto a 25x25 mm² acrylic glass slide. To minimize the adhesion of the crosslinked PGSA to the acrylic glass slide, the latter was treated with oxygen plasma. Multiple drop casting steps determined the final thickness of the film. In-between, the solvent was allowed to evaporate at 78°C or room temperature for several minutes. Crosslinking the individual as-cast layers for several minutes did not reveal superior film properties compared to simply letting the solvent evaporate before adding more PGSA solution. After reaching the desired thickness, removing as much solvent as possible in a vacuum oven before the final crosslinking step is essential to avoid the formation of bubbles and cavities in the PGSA. The viscous and slightly yellow film was put into a UV-irradiation chamber (LED Cube 100 IC, Honle UV technology, \( \lambda = 365 \text{ nm}, I = 600 \text{ mW/cm}^2 \)) for crosslinking. The time varied from 1 h to 7 h depending on the thickness and the concentration of DMPA in the film. During the process the temperature inside the chamber rose to approximately 50°C.\[^{[1,5]}\]
Other temporary substrate material besides oxygen-plasma treated acrylic glass were tested.

For the production of homogeneous films via drop casting, such a substrate should provide a low adhesion to the crosslinked film to ensure an easy removal after UV exposure and an even coverage with the PGSA solution. Neither glass nor its treatment with oxygen plasma, argon plasma, Cytop polymer, Zonyl surfactant or a pentafluorophenyltriethoxysilane self-assembling monolayer enabled a nondestructive removal of the cured PGSA film. On the contrary, the PGSA solution barely wet the surface of Teflon foil and formed big droplets. Argon plasma treatment of acrylic glass increased the adhesion of the PGSA and impeded an easy removal.

During the synthesis of one batch of PGSA with a 19% degree of acrylation, a prematurely crosslinked layer developed on the solution’s top surface during 2 weeks of storage in the fridge. This special 1.2 mm thick layer (25x25 mm² area) was used for tensile tests without further UV-induced crosslinking and is denoted as PGSA-19*.

S2.3. Embedded interconnect film fabrication with PGSA-19

A positive mold of the serpentine channels was laser-cut into multiple layers of cellulose acetate foil, which were stacked to 1 mm height. Then a negative cast mold was produced by pouring Ecoflex (by Smooth-On, 1:1 ratio) over the positive mold, creating serpentines of 1 mm height in a surrounding box. PGSA solution in EA (with added DMPA) was cast into the Ecoflex cast mold in multiple steps, interrupted by evaporation of the solvent first in a fume hood (to minimize bubbles) and then in a vacuum chamber, until a PGSA layer of 2 mm height covered the negative Ecoflex serpentines. Subsequent UV-curing took several hours because of the relatively large thickness (3.8 to 4.8 mm) of the PGSA film (25x25 mm² area) and the mold shielding the polymer from UV-light that is reflected from the sides or bottom of the chamber. The PGSA bottom layer with open serpentine interconnect channels could easily be removed from the cast mold.
Galinstan was filled into the serpentine channels with a syringe and then spread evenly with an ethanol-soaked pointy swab. The top layer was dropcast directly on top of the Galinstan-filled bottom layer (without a mold), again in multiple steps with intermediate solvent removal by drying and vacuum. Starting the dropcasting on the edge and moving in a circular pattern into the center was essential to avoid PGSA solution running down on the edge of the bottom layer. The straight interconnects for the LED and LEC pixels (1 mm high, 10 mm long, 2 mm wide) were fabricated in the same manner.

**S2.4. Thin film and embedded interconnect film fabrication with gelatin**

Different commercial products of gelatin were tested for thin films, each purchased from Sigma Aldrich: G2500 type A from porcine skin with gel strength 300 g Bloom, G1890 type A from porcine skin with gel strength 300 g Bloom for electrophoresis, 39465 from porcine skin for microbiology with ultrahigh gel strength. Samples in the main paper employ G2500 exclusively.

The gelatin films were produced by dissolving 6.65 g gelatin in 50 ml water under stirring at 50°C to 55°C until homogeneous and then adding 39.9 g glycerol to the solution in order to achieve a final mass ratio of water: glycerol: gelatin of 7.519: 6: 1. For tensile tests of thin film samples, the solution was dropcast to 3 mm thickness onto an acrylic glass or glass substrate (25x25 mm² area) and allowed to cool down to room temperature. The film could be easily peeled off the substrate.

For embedded interconnects, a negative mold of the serpentine interconnects was assembled from multiple layers of laser-cut cellulose acetate foil. These negatives were placed into a plastic box cast mold. Dropcasting the gelatin solution onto the negatives and removing the gelatin from the cast mold created the sample’s bottom layer with open interconnect serpentine channels (1 mm depth). These were filled with Galinstan by a syringe and with the help of an ethanol-soaked pointy swap. Directly dropcasting gelatin solution as a top layer did
not produce a reliable adhesion and caused delamination at low tensile stress. Instead, the top layer was cast separately in a plastic box. The adhesion of the top and bottom layer was promoted by the chemical reaction of two self-assembled monolayers. The top layer was treated with a 5 wt% solution of (3-aminopropyl)triethoxy-silane in ethanol by dropcasting, waiting for 10 min and rinsing the excess solution with isopropanol. The same procedure was conducted with (3-glycidyl-oxypropyl)trimethoxy-silane on the Galinstan-filled bottom layer. Both layers were connected and kept under pressure of a metal weight overnight to join the interfaces. The edge was fused by 15 min of oxygen plasma (final thickness 4.3 to 5.1 mm, 25x25 mm² area). The straight interconnects for the elbow mounted experiment (interconnect 1 mm high, 75 mm long, 3 mm wide), the LED and the LEC pixels (interconnect 1 mm high, 10 mm long, 2 mm wide) were fabricated in the same manner as the serpentine samples.

S2.5. Thin film fabrication of reference elastomers

For reference measurements, other elastomers were purchased as a film or processed into films of square shape (25x25 mm²). Ecoflex 00-20 was purchased from Smooth-On, mixed with a 1:1 component A:B ratio and cast into a plastic box cast mold (2.5 to 3.5 mm), removing bubbles in a vacuum oven at 60°C for 30 min. Poly(dimethylsiloxane) (PDMS, Sylgard 184) was purchased from Dow Corning, mixed with a 10:1 base-to-catalyst-ratio and cast onto an acrylic glass slide (1 mm thickness).

Sheets of vulcanized natural rubber of 300 µm thickness were used as purchased and cut to size (Späh NR Para 330 A Natur-Kautschuk). Formalate Latex moulding emulsion was used as purchased from Creartec and cast into a plastic box cast mold (0.6 mm thickness), accelerating the drying process at 60°C.

Terratek Flex (GDH-B1FA) was purchased from Green Dot Bioplastics and thermally flattened from grains into a thin film. After a pre-flattening step on a hotplate at 180°C, the final temperature during nanoimprinting (NIL CNI V1.0) for 1 h at 5.5 bar pressure between
two Teflon sheets determined the film thickness, ranging from 900 µm at 140°C to around 330 µm at 170 °C (Figure S5 a).

For mixed PEO-TMPE films, the linear poly(ethylene oxide) (PEO, $M_n = 5000$ kDa, Sigma Aldrich) and trimethylolpropane ethoxylate (TMPE, $M_n = 470$ Da, Sigma Aldrich) were mixed at various ratios in water or ethanol or water + 3 wt% ethanol at various concentrations, starting from a ratio of 12:1 and a concentration of 15 g·L⁻¹, as previously published. However, the resulting films failed our expectations, as they did not show any elastic properties but merely plastic deformation. The “optimized” high content of TMPE, as described in the publication, created a very oily film surface. Lower contents in TMPE created brittle & inelastic PEO films. Decreasing the amount of ethanol resulted in grainy solutions and films. Therefore, this material system was not investigated further and cannot be recommended as an elastic substrate.
S3. Mechanical properties of elastic materials

**Figure S5.** (a) Processing of Terratek Flex: Temperature-dependent thickness of the thermal flattening process. (b) Stress-strain curves for different products of gelatin. (c) Stress-strain curves of additional reference elastomers and PGSA-19*.

The mechanical properties of G2500 gelatin films, as used in the main paper, were compared to two other gelatin products (G1890 and 39465), using the same mass ratio of water:glycerol: gelatin = 7.519: 6: 1. As expected from the identical gel strength, G2500 and G1890 reveal very similar strain at break and tensile strength (**Figure S5 b**). The ultrahigh gel strength of gelatin 39465 results in lower values for both. Hence, the product G2500 was chosen for the samples with embedded interconnects.

Besides different types of gelatin, also different plasticizers and ratios of water:gelatin to plasticizer were examined. Lower contents in any plasticizer, e.g. 7.519:1:1 or 7.519:0.5:1, resulted in a hard and brittle film after drying and evaporation of the water. Alternative plasticizers to glycerol produced inferior film properties. TMPE (470 Da) created an oily surface, PEG (400 Da) created less elastic films and ethylene glycol inhibited the removal of the film from the carrier glass slide. Adding polymers instead of plasticizers, such as PEO (5000 kDa or 100 kDa) or PVA (85-124 kDa) did not produce elastic films.
Table S2. Mechanical properties of elastic materials.

| Material   | Young’s Modulus $E$ (kPa) | Ultimate Tensile Strength $\sigma_{UTS}$ (kPa) | Strain at break $\varepsilon_B$ |
|------------|---------------------------|----------------------------------------------|-------------------------------|
| PGS        | 143                       | 110                                          | 103%                          |
| PGSA-19    | 143                       | 81                                           | 130%                          |
| PGSA-19*   | 80                        | 265                                          | 360%                          |
| PGSA-28    | 592                       | 103                                          | 20%                           |
| Gelatin    | 48                        | 88                                           | 155%                          |
| Ecoflex    | 34                        | 161                                          | 522%                          |
| PDMS       | 1124                      | 2684                                         | 89%                           |
| Natural rubber | 2736                  | 7752                                         | 625%                          |
| Latex      | 866                       | 2190                                         | 708%                          |
| Terratek Flex | 2706                 | 523                                          | 58%                           |

Figure S6. Photographs of manually stretching biodegradable and not biodegradable elastic and not rubber-elastic materials.
S4. Surface-mounted interconnects

Figure S7. Surface-mounted Galinstan interconnects on PGSA-19*. (a) Tensile test: Stress and relative resistance of the interconnect versus strain. (b) Serpentine layout in (c) vertical position for tensile tests and (d) horizontal position for fatigue tests. (e) Fatigue test at 50% strain: Resistance for 1000 strain-relax cycles.

S4.1. Experimental

Galinstan traces on the surface of the elastomer material were a precursor to the embedded interconnect electrodes. They were patterned by stencil lithography, i.e., through a laser-cut (Trotec laserman) Teflon foil stencil (Bohlender, 120 µm thickness) on top of a sheet of PGSA-19* (1.2 mm thickness, 25x25 mm² area) in a straight, sinusoidal, semicircle and horseshoe pattern. Because of the adhesion of the PGSA-19*, no further fixation of the Teflon foil was needed and rather sharp edges of the interconnects could be achieved. Tensile tests with resistance measurements were conducted as described in the experimental part of the main paper. An in-house built setup was used for the fatigue tests, i.e., repetitive cycles of 50% strain at a strain rate of 1.06 mm·s⁻¹ with a simultaneous resistance measurement for
every cycle at full stretch and full relaxation. The resistance was measured with an Uno rev3 (Arduino) through a voltage divider.

S4.2. Results

Due to the vertical nature of the tensile test setup (Figure S7 c), the resistance of a surface trace rose over time, because Galinstan was slowly running to the bottom end of the trace. Thus, a change in initial absolute resistance could be observed, that increased with the number of measurements.

In stark contrast to the embedded traces, the resistance of the surface traces rose with increasing strain (Figure S7 a). The straight electrode showed the highest increase in resistance with rising strain by a factor 2.3 at 150% strain. It is the shortest electrode, but in comparison to the other patterns it lacks a “stretchability by design” feature and thus gets significantly thinner (Figure S7 b). The decrease in relative resistance of the sinusoidal and horseshoe serpentines is a result of the high initial resistance after accumulation of Galinstan on the bottom end of the trace and thinning at the top. For each pattern, the resistance displays a stronger hysteresis due to the accumulation of Galinstan at the bottom of the sample.

The horizontal fatigue test setup should weave the disadvantage of surface-mounted interconnects compared to embedded interconnects (Figure S7 d). During repetitive strain, the overall resistance decreases with an increasing number of cycles (Figure S7 e). This effect is likely caused by the removal of thickness bottlenecks in the Galinstan traces by evening the layer thickness. Secondly, the resistance in the stretched state is, as expected, higher in the beginning than in the relaxed state. Over time that changes and the resistance in the relaxed state exceeds the one of the stretched state. That might be explained with a continuous built up of oxide platelets on the surface that accumulate and reduce the cross-section of the relaxed state. In the stretched state, the accumulated oxide platelets distribute on the surface and increase the effective trace diameter.
Dropcasting a second layer of elastomer on top of the surface traces could protect them from smearing and oxidation. However, under strain, the Galinstan-filled channel contracted, permanently closed in multiple positions and separated the liquid Galinstan trace into bubbles. Hence, the route of creating pre-defined embedded channels was adopted, as described in section S2.
S5. Biodegradation of the LEC pixel

**Figure S8.** Biodegradation of PEDOT:PSS, the LEC stack and PGSA-19 with a cellulose reference, tested according to DIN EN ISO 14851 (2004-10): (a) Expressed as theoretical oxygen demand and (b) relative to the degradation of cellulose.

We proved the biodegradability of PGSA-19 in a standardized test by a plateau phase at 90% biodegradation (**Figure S8 a**). Relative to the cellulose reference, PGSA-19 reached a 62% relative degradation, which changes gradually after a steep initial rise (**Figure S8 b**). Besides the PGSA-19, also PEDOT:PSS and a full LEC stack were submitted to a biodegradation test. During this test, PEDOT:PSS and the blind control sample neither revealed a decrease in pH from 7.0 nor the development of larger flakes. The PEDOT:PSS test volume merely contained blue, finely suspended particles. This indicates the non-biodegradability of PEDOT:PSS in aqueous conditions, which is confirmed by absence of oxygen demand within 68 days (**Figure S8 a**). However, the stable pH indicates a bio-compatibility of PEDOT:PSS, which should be investigated further with toxicity tests.

The LEC sample used for the biodegradability test was identical to the stack of our previous publication.\cite{7} A cellulose acetate substrate was covered with a PEDOT:PSS cathode, a thin ZnO$_x$ hole injection layer, the active layer (super yellow, P(CL-TMC), TBABOB) and a PEDOT:PSS cathode. The cellulose acetate substrate comprised >99% of the volume and mass of the device. The cellulose acetate, the P(CL-TMC) ion-solvating polymer and the TBABOB salt were already certified to be biodegradable. The PEDOT:PSS and super yellow are likely bio-compatible, non-toxic and inert.\cite{8} ZnO$_x$ is a biocompatible substance, but
irrelevant for the biodegradation in terms of theoretical oxygen demand (ThOD). Therefore, while many of its components are biodegradable and the rest are inert, the entire LEC device may not necessarily be biodegradable.

During the biodegradation test of the LEC white flakes appeared, which are attributed to degrading biomass. Also, the decreasing pH value from 7.0 (blind control sample) to 6.9 also indicates biodegradation. A plateau value of 62% of the theoretical oxygen demand after 82 days corresponds to a 46% relative biodegradation compared to the cellulose reference (Figure S8 a, b). These results do not grant full biodegradability to the LEC stack, according to DIN EN 14995 and DIN EN 13432, but indicate partial degradation in aqueous media. Rather than the inert components of the stack, the slow degradation of cellulose acetate in water likely represents the limiting factor.

The stack used for the LEC pixel in Figure 5 differs from the aforementioned LEC stack by replacing the PEDOT:PSS cathode and ZnO$_x$ injection layer with a single inkjet-printed gold layer. The replacement of these two layers, which are irrelevant for biodegradation, with another biologically inert component should not change the biodegradability of the overall device. Hence, the data for the LEC stack in Figure S8 will apply equally to the LEC stack with the gold cathode.
S6. LIV characteristics under isotropic bending strain

**Figure S9.** Biodegradable LEC pixels mounted on (a-b) PGSA-19 and (c) gelatin elastic substrates with embedded Galinstan interconnects. (d) Swollen PGSA-19 substrate after printing the LEC layers directly on the elastomer. (e) Isotropic stretching of the elastic substrate with the mounted LEC at bending strains from 0% to 21%. LIV characteristics of LEC pixel interconnected by Galinstan on (f) PGSA-19 and (g) gelatin for different isotropic bending strains, with intermittent return to 0% strain.
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