

Imperceptible augmentation of living systems with organic bioelectronic fibres

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15 Abstract

Bioelectronics that provide functional and sensory augmentation for living structures, such as the human skin and plant epidermis, are enabling platforms for biology-machine interface, health management, and environmental monitoring. Ideally, such bioelectronic interfaces should not obstruct their hosts' inherent sensations and physiological changes. The full life cycles of the augmented living systems should also be designed to minimise environmental footprints. Here, we report imperceptible augmentation of living systems through in situ tethering of organic bioelectronic fibres. Using an orbital spinning technique, substrate-free and open fibre networks – based on poly (3,4-ethylenedioxythiophene) : polystyrene sulfonate (PEDOT:PSS) – can be tethered to morph biological surfaces, including fingertips, chick embryos, and plants. The customisable fibre networks enable biopotential sensing, skin-gated organic electrochemical transistors, augmented touch, and plant interfaces. We also show that the fibres can be used to couple prefabricated microelectronics and electronic textiles, while supporting on-demand device repair, upgrade, and recycle.

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Introduction

Merging biological systems with electronic devices could transform the way we interact and perceive our surroundings¹⁻⁹, providing, for example, data collection platforms for health management and environmental monitoring¹⁰⁻¹⁶. One goal in the development of functional and perceptual augmentation is to provide intimate bioelectronic device integration with living structures while minimally perturbing the biological functions of the host. Thin-film technologies^{14,16} can be used to create flexible electronics that conform to the macroscopic shape of biological surfaces, but their plastic substrates (around 3-10s micron thick) limit moisture/gas permeability. Electronic textiles^{6,17} use fibre materials or fibre-shaped devices and can offer enhanced comfort and breathability, but existing electronic textile fibre sizes are typically in the range of hundreds of microns, prohibiting intimate bio-integration.

Recent advances in stretchable electronics^{4,14,18}, electronic skins^{1,10,19}, nanomembranes^{3,4,20}, and nanomesh structures^{2,10,21} have led to augmentation technologies that are gas permeable^{2,3,10,20,21} and mechanically imperceptible to human skin^{1,10,19}. However, this level of imperceptibility could still be insufficient against the multi-faceted surface and bulk functions of living structures^{22,23}. In particular, biological pores, sensory receptors, and topography features²² can be concealed when films or components with limited openness are attached over large areas. Furthermore, the pressure exertion that is needed to transfer and deploy premade devices can preclude their use on deformation-sensitive surfaces.

The development of augmented living systems also needs to consider issues related to sustainability. Lithography-based microfabrication is energy and waste intensive due to the toxic chemical used, the need for sacrificial templates, and the effort involved in maintaining clean environments²⁴. The production and processing of traditional fibres and textiles also has large carbon and water footprints^{25,26}. In addition, the functionalisation of living structures that undergo dynamic transformations, or interact with biological analytes or chemical pollutants, can require regular full-scale renewal of the augmentation devices, which is environmentally costly.

Spiders build sophisticated fibre networks in situ, which are adapted to the environments and require minimal material consumption. Taking inspiration from spider webs, living structures could be augmented with bioelectronics based on designable open network architectures that use individual microscale fibres as building blocks. Such networks could be tethered onto living structures with tuneable fibre number density, orientation, and modalities (Fig. 1a). Three-

dimensional (3D) printing is considered an environmental-friendly fabrication route²⁸ that offers on demand fabrication²⁹⁻³². However, the resolution for state-of-the-art in situ printing is limited to hundreds of micrometres^{30,31}, which compromises device imperceptibility at the biological interfaces. On the other hand, existing approaches for fibre production – such as wet spinning³³,
5 melting spinning³⁴, or electrospinning³⁵ – can produce micro- and nano-fibres on a large scale, but lack advanced bioelectronic functions. Due to the low bending stiffness and low aerial footprint for surface adhesion of micro-scaled fibres, pre-functionalised fibre networks with open architectures are difficult to manipulate, and cannot be readily transferred and attached onto target objects³⁶. In situ generation of fibrous scaffolds is possible^{5,37}, but existing techniques
10 result in micro- and nano-meshes with random fibre overlays, and lack control in terms of fibrous patterns, surface contacts, and mesoscale network openness (Fig. 1b and Supplementary Table 1).

In this Article, we report the imperceptible augmentation of living structures through the in situ solution fibre tethering of poly (3,4-ethylenedioxythiophene):polystyrene sulfonate
15 (PEDOT:PSS)-based organic bioelectronic fibres. Our approach creates fibre interfaces that can be upgraded and repaired, and requires low material use and generate minimal waste (Fig. 1c). Our strategy could also extend the service duration of disposable and re-useable components, enhancing supply-chain resilience.

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In situ tethering organic bioelectronic fibre

Our organic bioelectronic fibres are based on PEDOT:PSS (a mixed ionic and electronic conducting polymer that have proven in vivo biocompatibility³⁸), hyaluronic acid (a skin extracellular matrix analogue³⁹ that helps fibre spinnability and skin contact), and polyethylene oxide. The bioelectronic fibres are produced from a solution phase at ambient conditions, where the solution spinnability could be characterised by the ratio of shear modulus over surface tension (Supplementary Fig. 7). We design an orbital spinning approach to control the bioelectronic fibre tethering and patterning directly onto living structures. Using the target periphery as a template, the fibre tethering is physically guided by the shape and position of the target object (Supplementary Fig. 8). Aided by the dynamic physically intelligent morphing mechanism of fibre tethering, in situ construction of fibre interface over a centimetre-sized target (e.g., a person's finger) does not require digital replica (Supplementary Video 1). In addition, the tethering process is tolerant to target movements for an electrode-patch application (Fig. 2a). Each rotating arm orbit results in one strand of solution fibre to be drawn onto the target. The fibre deposition path planning allows direct formation of fibre-to-surface and fibre-to-electrical contact connection (Supplementary Fig. 9). Thus, the entire fibre deposition process is contactless and mask-free. Controlling the fibre number density (*i.e.*, number of fibres (N) over the width of the fibre array (d), or N/d) allows tuning of the bulk optical property of the bioelectronic fibre patterns from transparent (transmittance ~ 98 %, for $(N/d) \sim 5/\text{mm}$) to semi-transparent (transmittance ~ 91 %, for $(N/d) \sim 20/\text{mm}$) (Fig. 2a and Supplementary Fig. 10). A summary of the bioelectronic fibre number densities used in various applications with the relationship to network opening and transparency is provided in Supplementary Table 2. The solution feeding consumes a total of ~ 1 μL solution per minute for direct fibre deposition. Considering typical fibre networks created within 2-5 minutes, the total solution usage is ~ 2 to 5 μL and the total embodied dry mass input could be estimated as 0.1-0.3 mg per fibre network device (Supplementary Note 1).

The fibre tethering process can take place on diverse biological objects with curved and irregular surfaces, from the width of a human hair, to ridges of a fingertip and chick embryos (Fig. 2b, Supplementary Fig. 11a-b). The bioelectronic fibre tethering process induces little perturbation to the targets' surface structures, where the force of a single fibre tethering is estimated to be in the range of 10 μN via cantilever experiments (Supplementary Note 2). For example, *Mimosa*

pubica, a touch sensitive plant⁴⁰ that closes upon gentle hand touch (force $\sim 200 \mu\text{N}$), does not respond to the fibre deposition process (Supplementary Video 2). The mechanical effects of fibre tethering on biocompatibility is further evaluated using fragile Day-2 chicken embryos, whose development is highly sensitive to external forces and stresses⁴¹. Our results show that the Day-2
5 chicken embryos with fibre networks on the developing tissue display normal growth rates and morphological changes through 24 hours post fibre tethering (Supplementary Video 3 and Supplementary Fig. 11c).

The fibres are spun in a solution/ wet state, meaning that abundant residual water remains in the ‘wet fibre’ upon surface tethering (Supplementary Note 3 & 4); thus, a dominant Wenzel-like
10 fibre-surface contact state is resulted. Furthermore, experimental results and theoretical analysis indicate that under the current spinning settings and formulation, intimate contacts over hundreds of microns of topographical features are expected to form on convex and solid structures (Supplementary note 4 and Supplementary Video 4). As shown in Fig. 2c, the bioelectronic fibre forms dominant intimate attachments even down to the micrometre-level surface topographies
15 for macroscopically convex surfaces (in contrast to non-contacting fibres produced by control solutions, Supplementary Fig. 12). Thus, depending on the contact states on different surfaces, the average feature size of a single bioelectronic fibre ranges between $1 \mu\text{m}$ and $5 \mu\text{m}$ (Supplementary Fig. 13). The spatially patterned bioelectronic fibres, along with their mechanical erasability in a wet state (as shown by fibre mechanical characterisation in
20 Supplementary Note 5), offer possibilities to create in situ patterning through both ‘additive’ and ‘subtractive’ modes (Fig. 2d, and further results later).

Imperceptible on-skin electrodes with tailored formats

A fresh fibre electrode on-a-fingertip (Fig. 3a), with contact impedance comparable to reported
25 microfabricated gold nanomeshes²¹, can be created within 3 minutes of fibre tethering (Fig. 3b and Supplementary Fig. 14a-b, under the current single nozzle setting). The interfacial contact impedance of fibre electrodes fabricated at different dates across a year fall within the range of around 20-40 $\text{k}\Omega$ at 1 kHz (Supplementary Fig. 14c). Such high success rates and consistency in deploying the fibre electrodes indicate that the functions of the fibre patch are negligibly affected
30 by positional drifts of the target during in situ fibre tethering. Figure. 3c and Supplementary Fig. 15 show that electrocardiogram (ECG) signals acquired by the bioelectronic fibre array are

highly consistent with the ECG signals collected by a reference gel electrode at the same time (Supplementary Video 5). Similarly, the fibre arrays could be configured to acquire electromyography (EMG) signals, and to monitor the steady increase of EMG signal amplitudes as representing the increased electrical activities of the skeleton muscles due to external loadings (Fig. 3d and Supplementary Fig. 16).

Repairability is a potential advantage of tethering the organic bioelectronic fibres as an exposed transient electrode. For example, as shown in Fig. 3e, when the exposed fibres were deliberately damaged, the fibre electrode-skin contact impedance at 1 kHz increases from around 20 k Ω to around 50 k Ω , subsequently affecting the ECG sensing performance (Supplementary Fig. 14e).

New fibres could then be deposited on-demand to repair the fibre electrode without affecting existing interconnections. The biopotential acquisition interface can then be fully renewed to recover the original contact impedance level and ECG sensing performance with a fraction of material inputs compared to creating a new electrode.

Next, we show that the device and contact formats of the bioelectronic fibres-on-a-fingertip can be customised to withstand various kinds of environmental and ‘touch’ perturbations simulating daily fingertip experiences (Supplementary Table 1). The tethered bioelectronic fibres, even in their fully exposed states, show stable electromechanical performance under various dry wearing conditions, and environmental disturbances such as water-soaking, humid, and mild heat (Fig. 3f). The specific conditions tested include (1) ambient wear for at least 6 hours; (2) more than 6,000 times of mouse clicking with a mean clicking force of ~ 1.5 N; (3) around 25 meters of dry frictional wear with a plastic surface with a mean normal force up to 3 N; (4) under the simulated ‘wet’ or ‘heat’ conditions without mechanical disturbance (*i.e.*, for at least 30 minutes either immersed in water, or in 90 % relative humidity, or under ~ 40 °C environment) (Fig. 3f, Supplementary Fig. 17, and Supplementary Video 6). Under these conditions, the on-skin fibre patterns exhibited no visible macroscopic distortion, and there are insignificant performance degradations in terms of interfacial contact impedance and ECG acquisition. The conformally attached bioelectronic fibres form good fibre-to-skin adhesion, with the maximum recorded peeling force approaching around 15 N/m (Supplementary Fig. 18). It is to note that the strength, and thus the electromechanical performance of the bioelectronic fibres, are affected by the level of fibre hydration as shown in Supplementary Note 5. Therefore, under wet mechanical disturbances (*i.e.*, water rinsing), the exposed bioelectronic fibres on the fingertip become

unstable. Further enhancement in the ‘wet-stability’ of the device interfaces can be designed through incorporating biocompatible and biodegradable cellulose-based materials as protective layers. As a conceptual demonstration, as shown in Fig. 3g and Supplementary Fig. 19, cellulose-based fibres can be added on top of the bioelectronic fibres, to improve the overall fibre device’s electromechanical stability. With the cellulose-based protective layers, the tethered bioelectronic fibre array can maintain its as-deposited performance for ~ 8 meters of wet friction with a normal force of ~ 0.5 N, and at least an hour of computer typing and office work. Furthermore, the exposed fibre contact (*e.g.*, the contact connection between the bioelectronic fibres with the copper tape on the nail) could be encapsulated by a cellulose-based film of around 2 μm thickness. In this case, the entire fibre device on the fingertip could withstand at least 10 cycles of 30 seconds running water rinsing (Fig. 3h and Supplementary Fig. 20).

Substrate-free fibres for imperceptible augmentation

We demonstrate concepts of how customising bioelectronic fibre patterns could offer myriad possibilities for imperceptibly augmented living structures. First, as both sides of the bioelectronic fibres can remain exposed when worn on the finger, the wearer (person-i) can detect another individual (person-ii)’s ECG by contacting the wearable electrode with the other person (person-ii)’s bare finger or wrist (Fig. 4a). The dual-ECG signals acquired by the fibre electrodes contain ECG characteristics of the two people: the R peaks of person-i are pointing upwards because the person-i ECG is measured from the left to right hands; while the R peaks of person-ii are pointing downwards because it is measured from right to left hands (*i.e.*, in the reverse direction compared to person-i). A signal processing algorithm could identify the upward and downward R peaks for both people (Supplementary Fig. 21). It is to note that the dual-ECG signals measured from the fibre electrodes show a high correlation coefficient ($P = 0.94$) with the reconstructed composite-ECG signal measured from individual’s validation gel electrodes. In the future, advanced signal processing techniques, including machine learning and blind signal separation⁴², could be used for ECG signal separation and identification of other minor peaks from the dual-ECG signals. Because the fibre arrays are substrate-free, and the open fibre network minimally conceal the skin surfaces, the subtle touch sensations of the volunteers are preserved so that they can simultaneously feel the blood vessel pulsations underneath the skin. In addition, the semiconducting nature of PEDOT:PSS³⁸ offers the opportunity for it to be

configured into an organic electrochemical transistor (OECT). Bioelectronic fibres are tethered to form a breathable skin-gated OECT on the fingertip, where the area of skin acts as electrolyte between the gate and the substrate-free channel fibre arrays. The conformal contact facilitates charge exchange at the skin-fibre interface, and this enables the gating of OECT using skin as the gate-channel electrolyte (Fig. 4b and Supplementary Note 6). The applied channel voltage at 30 s generates a positive current in the fibre array, then the current drops abruptly as expected for PEDOT:PSS channel material operating in depletion mode (*i.e.*, positive gate voltage switching off the device, and vice versa). The removal of the gate voltage at 90 s leads to the recovery of the current (hence the recovery of the channel conductance). Repeated gate voltage pulses result in similar current responses, showing the fibre array remains structurally intact during the switching processes.

Furthermore, complementing bioelectronic fibres with other fibres of different sensing modalities at the same anatomical site offers the opportunities to create multi-modal sensors. Humans do not possess skin ‘wetness’ receptors, and ‘wetness’ is interpreted individually through perceptions of temperature and mechanical inputs⁴³. The resistance of PEDOT:PSS materials is moisture dependent⁴⁴. As an intuitive illustration, Supplementary Video 6 shows that bioelectronic fibres tethered on a dandelion seedhead could be used to detect the environmental moisture flow without concealing the seedhead’s fine hairs. As a conceptual demonstration of augmented mist pulse perception without interfering with the host’s intrinsic perception, bioelectronic fibres and colorimetric pH-responsive fibres are both looped on the index finger of a person (Fig. 4c, see Materials and Methods for the fabrication of pH-responsive fibres). The temporal resistance of bioelectronic fibres would increase upon water mist pulses. Impingement of acidic, neutral, or alkaline mist pulses onto the finger can be distinguished by simultaneously monitoring the bioelectronic fibres’ temporal resistance and the pH-responsive fibres’ colour. Dual-modal sensing (mist detection by bioelectronic fibres, and pH by colorimetric fibres) is used here, because if mists of similar pH repeatedly impinge on the fingertip, the bioelectronic fibres will indicate the mist flows by the changes in electrical resistance, but the colorimetric fibres’ colour will remain the same. Because the fibre arrays are substrate-free and minimally conceal the skin surfaces, all volunteers were able to feel the subtle sensations generated by the mist flow impingements through the fibre arrays.

Adaptive and reconfigurable fibre sensing arrays and networks

We further demonstrate adaptive and reconfigurable sensing systems based on the bioelectronic fibres (as the sensing elements), coupled with prefabricated microelectronics or e-textile wearables. The ability to control the fibre orientations (θ) enable versatile inter-connections to be made into a device. Parallel ($\theta=0^\circ$), parallelogram ($\theta=\pm 15^\circ$), and fanning ($-30^\circ < \theta < 30^\circ$) patterns have been used for various applications in this work. The estimated patterning precisions were shown to be all above $\sim 75\%$, as indicated in Fig. 5a. The patterning precision estimations are affected by the orbital spinning process-intrinsic factors including mechanical controls, and environmental disturbances (*i.e.*, wind) during patterning; but could also be a result of mis-identification due to automatic image registration of the fibres during post data analysis. Individual fibres' continuity and form factor thus support the connection of small electronic devices such as a micro- light emitting diode (LED) without adhesives (Fig. 5b). The low deposition forces imposed by the orbital spinning and fibre tethering (Supplementary Note 2) mean that the micro-LED could stay still by gravity and by the friction of the leaf texture during the circuit formation. In the case of the micro-LED used, its weight is $\sim 1.4 \times 10^{-5}$ N. Thus, considering the failure force per fibre during debonding is $\sim 3.5 \times 10^{-5}$ N from Supplementary Fig. 17, as few as a single fibre is sufficient to support the weight of one micro-LED. Cyclic voltammetry through the bioelectronic fibres showed that they display ohmic resistance under up to 6V/cm applied voltage (Supplementary Fig. 23), making them compatible with other low-power bio-safe electronic components. In the subsequent demonstration, distributed bioelectronic fibres on a plant could be used to connect with the micro-LED, as a display, to form a warning system for elevated levels of ammonia exposure (Fig. 5c). Ammonia is a type of n-type dopants that interacts with PEDOT:PSS in the bioelectronic fibres to cause a de-doping of the PEDOT:PSS polymer backbone⁴⁵; hence, the micro-LED dims non-reversibly upon ammonia exposure (Supplementary Fig. 22a). In comparison, the micro-LED light would only dim temporarily when encountering water mist because the effect of water on the resistance of PEDOT:PSS is reversible (Supplementary Fig. 22b and Supplementary Video 7). The designed bioelectronic fibre pattern widens the ammonia mist capture area without compromising breathability and light transmission (*e.g.*, over 90 % transmittance as shown in Fig. 2a) for photosynthesis of the leaf surface. The fibre array, which acts as a transient interface, can be renewed independent of the re-usable LED (or other discrete electronic components). The fibre

arrays, after being pulled off from the leaf, could be recycled through grinding and sonication, to produce a conducting fibre-loaded ink for 3D printing (Fig. 5d).

In the third demonstration, we show a rewritable and reconfigurable fibre array and network on a leaf of a whole plant (Fig. 5e). The fibre tethering could enable a “fabrication closed-loop” of “writing, erasing, overlaying” (Fig. 5e-i) for in situ sensing interface reconfiguration and renewal. “Writing” is an additive process which involves deploying fibres in the target area; “erasing” selectively removes fibres, where the weakened strength of bioelectronic fibres in wet regions enable them to be selectively erased off on-demand without needing organic solvents; and “overlay” (as an additive process) deploys fibres over existing structures with an arbitrary alignment. When reconfiguring the fibre path for chemiresistive sensing, customising the number of fibres (N) in the array could enable the circuitry level fibre array resistance to fall within a desirable range (Supplementary Fig. 24). This conceptual reconfigurable sensing interface could be advantageous where if the leaf surface is damaged or obstructed, then the bioelectronic fibres could be ‘renewed’ by re-routing the fibre path onto the original electrical contact connections without further perturbing the living structure (leaf) (Supplementary Fig. 24). Figure. 5e-ii shows that such mask-free direct patterning supports in situ sensing interface repair and reconstruction on living structures with minimal disruption and infinitesimal material usage.

Finally, we demonstrate interface compatibility between the fibre tethering strategy and e-textile wearables (Fig. 5f). Here, bioelectronic fibres are tethered directly onto a glove sewn with metallic conductive yarns. Such tethering provides a dry interfacial coupling which drastically decreases the contact impedance between the metallic yarn of the glove and human skins, enabling biopotential sensing through touch. Afterwards, the bioelectronic fibres, which are coupled to the e-textile through a dry mechanical interface, could be removed from the glove by dry scratching. The collected bioelectronic fibres could also be recycled for 3D printing as shown in Fig. 5d. Thus, bioelectronic fibre tethering-enabled augmentation can be considered a sustainable bridging technology, as it offers the possibility to decouple the service durations of disposable, and multi-use, quasi-permanent components.

Conclusion

We have reported the augmentation of living structures through the in situ tethering of organic bioelectronic fibres. The imperceptible fibres are fabricated on demand and can adapt to the living structures without influencing their biological functions and transformations. By harnessing the viscoelasticity and surface wetting properties of the pre-dry solution during in situ fibre tethering, we create sensing interfaces across biological curvatures and topographies of different scales (such as fingertips and finger ridges). The in situ fibre tethering approach overcomes material and format limitations associated with prefabricated interfaces. Added by the physical intelligence morphing mechanism, the fibre tethering process lowers the requirements for intrinsic material stretchability and complex print path planning, while enabling rigid-to-flexible device coupling (such as coupling microelectronics to plant leaves).

Currently, the stretchability of the bioelectronic fibres is limited by the intrinsic material properties of the PEDOT:PSS and PEO. Nevertheless, by modifying the fibre network design patterns and orientations with respect to the stretching direction, the device interface cyclic stretchability can be enhanced to around 15 % (Supplementary Fig. 25). In the future, the stretchability of the fibres could be enhanced further by combining with elastomers. Furthermore, the functionality and stability of the resulting biointerfaces could be tailored by mixing-and-matching a wide range of fibre materials (or fibre modalities).

Today, developments in electronics and sensors need to be focused on more than just increasing device performance — they also need to consider ways to reduce environmental impacts over the full life cycle of the devices^{24, 46–48}. The raw materials used to fabricate our organic bioelectronic fibres, and their assembled device interfaces, are based on earth-abundant and biocompatible materials (including organic semi-conductors PEDOT:PSS, and cellulose derivatives), and are not reliant on precious metals or supply-chain sensitive sources. Our fibre tethering approach also offers an individually-adaptive fabrication strategy with low energy consumption (Supplementary Note 7). The 0.1–0.3 mg of dry mass input that is required to form our fibre networks for each device is equivalent to the estimated microfibre mass released from 1 gram of synthetic fabric after machine washing⁴⁹. A typical machine washing cycle of 5 kg of fabric generates a greater environmental cost in terms of water consumption and microparticle production than fabricating 5,000 bioelectronic fibre arrays (Supplementary Note 8). Our material and process strategy for bioelectronic fibre tethering — which includes creation, repair,

reconfiguration and recycle steps — thus offers a low material and low energy consumption approach to augmenting living systems with minimal environmental impact.

Methods

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Solution preparation

Bioelectronic fibres were prepared by mixing a PEDOT:PSS (poly (3,4-ethylenedioxythiophene) : polystyrene sulfonate) solution and a PEO (polyethylene glycol)/ HA (sodium hyaluronate) solution. The PEDOT:PSS solution was prepared according to the literature for achieving good conductivity and stability⁵⁰. 95 % (v/v) of PEDOT:PSS solution (Heraeus Clevis PH 1000, as ~1 % (w/w) aqueous dispersion) was mixed with 5 % (v/v) of ethylene glycol (Sigma-Aldrich), and additional 10 μ L of DBSA (dodecylbenzenesulfonic acid, Sigma-Aldrich) was added to per 10 mL of the solution as a surfactant to prevent aggregation. The solution was then sonicated for 20 min. The PEO/ HA solution was prepared by dissolving 2 % (w/w) 8 M Da PEO and 0.5 % (w/w) HA (Sigma-Aldrich) in deionised water by mild stirring at room temperatures for 48 hours. The choice of PEO was based on our previous fibre printing experience³³, where we demonstrated that the long molecular chain PEO solutions (4 M Da) could be stretched into thin fibres without the need of electrical field. In this work, we used an even higher molecular weight PEO (8 M Da) along with HA to further enhance the fibre spinnability and fibre interfacial contacts. Before fibre deposition, the PEDOT:PSS solution and PEO solution were mixed together in 2:1 (v/v) ratio for achieving good fibre spinnability with sufficient PEDOT:PSS for sensing applications, and then it was stirred for 12 hours at room temperatures to form the final bioelectronic fibre solution.

The solution for forming the protective cellulose-based fibre layers was prepared by dissolving 6 % (w/w) of Ethyl Cellulose and 1.5 % (w/w) of 8 M Da PEO (Sigma-Aldrich) in 80 % (v/v) ethanol, followed by stirring at room temperature for 12 hours. This materials concentration was adapted from previous literature⁵¹, and 8 M Da PEO was used in this work to enhance the fibre spinnability.

In the case of dual-modal sensing on the fingertips, a fully aqueous PEO-based fibre solution was used for skin-compatible pH-sensing. To prepare the PEO-base pH-responsive fibre solution, 6 % (v/v) nitrazine yellow (a pH-responsive dye, Sigma-Aldrich, of which

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concentration allows visible fibre colour change with good spinnability) was added to the PEO base matrix solution for mild stirring at room temperature for 12 hours.

Fibre tethering process

For the current study, the orbital spinning platform for fibre tethering consists of a cylindrical fibre spinning zone of up to 15 cm in diameter and 30 cm depth. The rotating arm was powered by a servo motor (Parallax 6 V continuous servo) at the rate of ~ 45-65 rpm. The diameter of the spinning zone can be adjusted depending on the size of the target object, and could be varied from 0.5 to 15 cm. The solution feeding system and the rotating arm formed the fibre tethering platform, and the platform could be mounted on a translational stage, a mini-rover or being handheld. The fibre solution was loaded into a 1 mL syringe that was connected to a 0.25 inch long 22-gauge blunt-end stainless steel needle (Adhesive Dispensing Ltd). The syringe and the needle were placed above the rotating arm in the way that the pendent solution droplet at the tip of the needle would just be scratched by the rotating arm. The fibre deposition path is designed such that in each cycle of fibre tethering, the fibre is tethered onto the target surface (e.g., the fingertip), as well as the contact electrode (e.g., copper tape on the nail) in a single orbital spinning step. The contact electrodes made of copper tapes then connect to external measuring instrument.

For lab-based experiments, pressurised air (Elveflow OB1 microfluidic flow controller) was connected to the syringe to feed the solution (~ 40 mbar, flow rate ~ 60 $\mu\text{L}/\text{min}$). The linear movement, controlled by a linear translational stage (Thorlabs MTS50-Z8), was responsible for creating parallel fibre arrays with various densities; and the rotational movement, controlled by a servo motor (20 kg high torque servo, SUNFOUNDER), was responsible for creating fibre arrays with various angles.

Fibre solution rheological characterisations

The shear rheological properties of various fibre solutions were acquired with a Kinexus KNX2112 controlled stress rheometer at 25 °C. A parallel plate configuration was used with 1 mm gap distance. Fibre solution extension measurements were performed with TriMaster. The extension speed was set at 80 mm/s and the images were acquired by a PHANTOM VEO-E 310L high-speed camera at 3,000 frames per second.

The surface tension of the solutions was determined through pendent drop shape analysis⁵². Solutions were fed steadily (flow rate 300 $\mu\text{L}/\text{hr}$) into the outlet of a stainless steel

needle (18 gauge, the outer diameter is 1.8 mm). The photos of the droplets were taken just before the droplets were to break off from the needle, and they were analysed through Pendent_Drop plugin by ImageJ (imagej.net/plugins/pendent-drop).

In the experiment to investigate local fibre contact status with target surfaces, a glass capillary with outer diameter of 675 μm was placed on the surface of a microscopic glass slide (length 76 mm), and then a fibre was deposited onto the glass slide and the glass capillary. The fibre spinning and wetting process was in situ recorded. The glass slide was either treated to be hydrophilic (water contact angle $\approx 0^\circ$) or hydrophobic (water contact angle $\approx 90^\circ$), through 30 minutes of UV plasma treating or 20 seconds of Sigmacote soaking, respectively.

10 **Biocompatibility tests with chicken embryos**

Wild type fertilized chicken eggs, acquired from MedEgg Inc. Eggs, were stored in a monitored 15 $^\circ\text{C}$ fridge and incubated at 37.5 $^\circ\text{C}$ and $\sim 45\%$ humidity in egg incubators (Brinsea). The embryo extraction and ex ovo culture were performed as previously described⁵³. PEDOT:PSS-based bioelectronic fibres were used for the chicken embryo experiments.

15 **Fabrication of fibre biopotential monitoring electrodes**

For contact impedance and ECG measurements, a strip of copper tape with a wire to serve as connections to the outer circuit was first placed onto the nail of the index fingers of both hands of the volunteer. The PEDOT:PSS-based bioelectric fibres were then deposited onto both index fingers (left hand index finger first and then right hand) of the volunteer for the measurements of contact impedance and ECG signals. An array of parallel bioelectronic fibres (an array of 180 parallel fibres across 9 mm, $\frac{N}{d} = \frac{180}{9\text{ mm}}$) were used. To create such an electrode, during fibre spinning, the translational stage was set to move at a constant speed to deposit the fibre array to loop around the fingertip and the copper electrode on the nail. The interfacial impedance between the fibre and the skin decreases as the number of fibres N increases approx. linearly with time (see discussion notes in Supplementary Fig. 14). For this demonstration, the translation stage speed was set at $\sim 50\ \mu\text{m/s}$, thus a total distance of $\sim 9\ \text{mm}$ was covered over a $\sim 3\ \text{min}$ deposition time. Fibres on the left hand index finger was used as the working electrode, and fibres on the right hand index finger was used as the counter electrode. For EMG measurements, a strip of Kapton tape was first placed above the metacarpal bone of the thumb for insulation purpose, and then a strip of copper tape with wire was then placed on top of the Kapton tape. Fibres were then deposited onto thumb-thenar muscle region. An array of parallel

bioelectronic fibres (an array of 360 parallel fibres across around 2 cm, $\frac{N}{d} \sim \frac{360}{2 \text{ cm}}$) were used, and during deposition, the fibre spinning platform moves linearly at a constant speed of 50 $\mu\text{m/s}$ to allow fibre deposition covering the muscle area. The loading of various weights was applied to the thumb.

5 The impedance was measured by a PalmSens4 Potentiostat; and the ECG and EMG signals were measured with Intan RHS Stim/Recording System. To compare the ECG signals obtained from the fibre and gel electrodes, both sets of ECG signals were simultaneously collected from the same individual. For working electrodes on the lefthand, the fibre electrode was positioned on the fingertip of the index finger, and the gel electrode was placed on the
10 fingertip of the middle finger. Additionally, a separate fibre electrode was placed to the fingertip of the righthand index finger as counter electrode. The signals exhibited high consistency with the correlation P values range from ~ 0.95 to 0.99 after applying the signal filtering. The ECG signals were filtered through a band-pass filter of 0.5-50 Hz, and EMG signals were filtered through a band-pass filter of 0.5-500 Hz. The gel electrodes used as references were commercial
15 disposable ECG Electrodes (ADInstruments, Oxford). The Pearson correlation coefficient (P) between ECG signals was calculated by MATLAB.

Fibre electrode stability and repairability study

 The surface debonding/ peeling experiment was performed by depositing an array of ~ 500 bioelectronic fibres onto a cherry tree leaf. Both ends of the bioelectronic fibres are attached
20 and glued onto two parallel lifting arms, which lift the fibre array to peel it off from the leaf at a constant speed of 50 $\mu\text{m/s}$. The force is measured by a balance (Ohaus Scout Portable Balance, 120 g Capacity, resolution 0.001 g) at 1 reading per second. It is to note that the ASTM D2861 test (90 degree peeling test) was not replicated here due to the difficulties of manipulating the thin substrate-free fibre arrays on a plant leaf.

25 In the mouse clicking experiment, and the clicking force of the mouse (Onecall, CS33211) was measured by a force gauge (RS Component, FK 50, resolution 0.02 N). The mouse clicking speed is around 1 click per second. In the dry friction experiment, a cylindrical roller (outer diameter 2 cm) was 3D printed with polylactic acid or polylactide (PLA) material and mounted on a servo motor (Parallax 6 V continuous servo, ~ 40 rpm). The speed of friction
30 between the roller surface and the fingertip is ~ 4 cm/s. The whole set-up was placed on top of a balance (Fisher Scientific, CSC 5000, resolution 1g) in order to measure the normal pressing

force. Dry friction experiments were performed in an air-conditioned room to minimise perspiration. The same experimental set-up was used in the wet friction experiments, DI water was sprayed (~ 0.2 mL) onto the fibre electrodes on the fingertip for every 30 s (\sim every 120 cm friction distance). The cellulose-based protective fibre layer (an array of 90 parallel fibres across ~ 9 mm, $\frac{N}{d} \sim \frac{90}{9\text{ mm}}$) were deposited on top of the bioelectronic fibres at an angle of $\sim 30^\circ$. Hand rinsing was performed with running tap water. In each rinsing cycle, the fingertip side was directly faced with running water for 10 seconds, and then the nail side (with the fibre contact) was directly faced with running water for 10 seconds; this is followed by 10 seconds resting until the next rinsing cycle. The bioelectronic fibres were protected with a layer of cellulose-based fibres of an array of 90 parallel fibres across ~ 9 mm, $\frac{N}{d} \sim \frac{90}{9\text{ mm}}$, and then the fibre connection area on the nail was encapsulated by applying a cellulose-based liquid plaster film (nitrocellulose as the main solid ingredient, New-skin Liquid Bandage Spray).

Augmented touch

For the case of touch dual-ECG sensing, an array of bioelectronic fibres (180 parallel fibres across 3 mm, $\frac{N}{d} = \frac{180}{3\text{ mm}}$) were deposited onto the index fingers of person-i. During the measurements, bioelectronic fibres are tethered onto both the left and right hand index fingers of person-i to serve as working and counter electrodes respectively. Then person-i would touch person-ii's index finger or wrist area so that the fibres would be in direct contact with the skins of both people. Individual ECG signals of both people for validation were acquired as references using commercial disposable ECG gel electrodes at the same time (ADInstruments, Oxford). The raw ECG signals are measured by the gel electrodes attached to individual persons. The purpose of the individual ECG signal measured by gel electrodes (Supplementary Fig. 18a) is to validate that the signals measured from the fibres could reflect the ECG of both people. The reconstructed dual-ECG signal in Fig. 4a is obtained by superimposing the individual ECG signals measured by the gel electrodes (with a scaling factor of 0.5).

Dual-model sensing

An array of bioelectronic fibres (~ 100 fibres parallel fibres across 0.5 cm) and an array of PEO-base pH-responsive fibres (~ 400 fibres parallel fibres across 0.5 cm) were looped onto the tip of the index finger separately. Two strips of copper tapes were placed on the nail to serve as electrical connections to measure the resistance change of the bioelectronic fibres. The

resistance changes of the bioelectronic fibres, upon applying mists, were acquired by a multimeter (Keysight 34465A). An ultrasonic humidifier (VicTsing Essential Oil Diffuser) was used to generate humid mists of various pH levels. Acidic mist was generated from a solution by adding 100 μ L white vinegar into to every 10 ml of water (pH of the solution \sim 3); alkaline mist was generated from a solution by adding 0.5 gram of laundry powder (Non-bio laundry powder, Sainsbury's) into every 10 ml of water (pH of the solution \sim 12).

Skin-gated organic electrochemical transistor (OECT)

The OECT measurement was commenced using a standard Source Measurement Unit (SMU, 2612B, Keithley). Fibre arrays were deposited on the tip of a little finger. One SMU channel was connected through the fibre array channel to its counter electrode, and the other SMU channel was connected to a gate electrode through a single end connection (schematically shown in Supplementary Fig. 6). Both SMU channels were configured to share the common ground and have a constant current limit at 1 mA. The channel voltage was set at 1 V, and the gate voltage was pulsed between 0 to 0.5V.

UV/Vis characterisations

The UV/Vis spectra were acquired with a JENWAY 7250 UV/Vis Spectrophotometer by depositing fibre arrays of various densities on glass cover slips.

Plant interfaces

In the experiment of environmental ammonia sensing for plants, an orange LED (ROHM Semiconductor, SML-811D TT86A) was placed onto the leaf of an orchid plant with a tweezer (without any adhesives), this was followed by orbital spinning of bioelectronic fibres that selectively 'wraps' around the electrode contacts of the LED light along with the parts of the leaf surface (\sim 75 fibres in each array, and the length of fibres \sim 4 cm in each array). The fibre tethering would provide both mechanical mounting support and electrical connections for the LED. In this case, the fibre arrays were deposited in a fanning pattern (fanning angle of \sim 15°, $\frac{N}{\theta} \sim \frac{75}{15^\circ}$ for each electrode contact) with the centre on the electrodes of the LED. The system was powered by a DC voltage supply (Digital Bench Power Supply 180 W, RS Component), and the driving voltage (\sim 5 – 13 V depending on fibre array resistance, the electrical field does not exceed 6 V/cm in the fibre array) was tuned to just light up (turn on) the LED light prior to ammonia exposure (see detailed discussions on the operation of the warning system in

Supplementary Fig. 22). Ammonia solution (2.3 %) was sprayed onto the leaf of the plant to simulate the environmental ammonia contamination. The resistance change of the fibre array as a result of ammonia contact would result in a change in LED brightness (which is recorded through a video). The bioelectronic fibres' resistance in response to varied concentrations of ammonia vapour was tested an array of fibres with $\frac{N}{d} \sim \frac{75}{3 \text{ mm}}$ and 20 mm length on a microscope glass slide. The fibres, on the bottom surface of the glass slide, were placed above the ammonia solutions of various concentrations (1.7, 2.3, 2.8 % in water) at a distance of 2 cm for 2 minutes.

Reconfigurable fibre sensing interface

The steps of creating a reconfigurable fibre sensing interface on an orchid leaf is shown in Supplementary Fig. 24b. First, two orange LEDs (ROHM Semiconductor, SML-811DTT86A) were placed on the surface of an orchid leaf, followed by fibre tethering. Four arrays of bioelectronic fibres (~ 60 fibres in each array with $\frac{N}{d} \sim \frac{60}{1 \text{ mm}}$) were deposited to connect the electrodes of the LEDs respectively (step 1&2). Under this configuration, only one LED could be powered under one power supply channel. In the reconfiguration process (step 3&4), a cotton bud with 70 % ethanol was used to selectively erase unwanted fibre array traces, followed by tethering new fibre arrays to form a new sensing interface (the number of fibre N might need to be adjusted depending on the reconfigured fibre path, see additional discussion in Supplementary Fig. 22 & 24). Under the reconfigured circuit, both LEDs could be powered without need to change the outer circuit connections. A power supply was used (Digital Bench Power Supply 180 W, RS Component) to provide DC voltage ($< 13 \text{ V}$) in this experiment.

Fibre tethering with e-textile

Stainless steel yarns (Rapid Electronics, 87-6102, Light Stitches Conductive Thread Kit) were sewed to cover the fingertip region of white cotton gloves, followed by bioelectronic fibre tethering on top of the stainless steel yarn region (180 fibres across around 5 mm distance, $\frac{N}{d} \sim \frac{180}{5 \text{ mm}}$). During the biopotential measurement, the person wearing the glove put the finger onto the wrist area of the other person, with the fibre coated region directly in contact with the lefthand wrist of the other person. A hydrogel pad was connected to righthand wrist of the other person to serve as the counter electrode. The stainless steel yarns were connected to the working electrode of an electrophysiology machine (Intan RHS Stim/Recording System) for ECG measurement, and to a Potentiostat (PalmSens4) for contact impedance measurement. After

measurement, the bioelectronic fibres could be removed from the glove by brushing and recycled.

Recycle of wasted bioelectronic fibres

The bioelectronic fibres could be removed from the target objects (*i.e.*, leaf and e-textile glove) after using, and then could be recycled for making conducting 3D printing inks. The recycled fibres, in dry status, could be redispersed into 70 % ethanol (also acts as a sterilisation medium) at a concentration of 1.5 % w/w, 1 % of 400 k Da PEO powders were also added. Then the dispersion was grinded for 5 minutes to form a homogeneous solution paste, which could be loaded into a syringe for printing.

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Data availability

All source data (<https://zenodo.org/records/10808385>) are available in the main text and supplementary information; any additional information needed could be requested from the corresponding author.

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Author contributions statement

W.W. and Y.Y.S.H. conceived, designed experiments, and performed analysis. W.W. devised the fibre spinning set-ups and performed fibre device fabrications and characterisations, and biopotential measurements. Y.P. optimised OECT and devised the reconfigurable fibre fabrications. Y.S. performed pH fibre sensing experiments. I.M.L assisted general data analysis and S.G.S.K. assisted fibre durability and characterisations. S.V-B., Yang. C., S.B.P.M., and Y. C. assisted testing and characterisations. T.S. proposed theoretical analysis for fibre wetting. Y.Y.S.H. supervised the project, and T.H., F.X., and G.G.M. advised the project. All authors discussed results. W.W. and Y.Y.S.H. wrote the manuscript with helps from all authors.

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Ethics statement

Human participant experiments were performed with the approval of the Ethics Committee of the Department of Engineering at the University of Cambridge (7/7/2021, CUEDREC) and after obtaining informed consent from volunteers. No animal protocol was required for the chicken embryonic stages studied (< 2 weeks) under the UK Animals (Scientific Procedures) Act 1986.

Competing interests statement

University of Cambridge has filed a patent application related to this technology. The patent application number is GB2307449.5 (by YYSH, WW, YP and YC). The authors declare that they have no other competing interests.

Figure captions:

Figure 1. Imperceptibly augmented living structures with organic bioelectronic fibres. **a, (i)** fibre number density (number of fibres (N) over the width of the fibre array (d)), fibre orientation (θ), and modalities (fibre materials and pattern design) can be customised during in situ fibre tethering; **(ii)** intimate contacts are achieved between the organic bioelectronic fibres (each $\sim 1\text{-}5\ \mu\text{m}$ wide) and different biological surfaces with micro to millimetre-scaled topographies; **(iii)** the bioelectronic fibre arrays are reconfigurable to support scalable customisation of electronic and sensing elements on living structures in situ. **b,** Length scales/ feature sizes are indicated for **(i)** biological structures on the human skin of a hand, including sweat pores (density $\sim 250\text{-}500/\text{cm}^2$, symbol P1; pore size $\sim 60\text{-}80\ \mu\text{m}$, symbol P2⁵⁴), fingerprint ridges (millimetre ridge-to-ridge spacing, symbol F1; and ridge height $\sim 20\text{-}40\ \mu\text{m}$, symbol F2⁵⁵), single skin cells (sizes $\sim 30\ \mu\text{m}$, symbol C⁵⁶), and receptor fields on the fingertip (\sim millimetre range, symbol R⁵⁷); **(ii)** bioelectronic fibre tethering for its fibre width, thickness and network opening; **(iii)** nanomesh for its mesh thickness and mesh opening^{2,5,21}. **(iv)** in situ printing or thermally-drawn e-fibres for their line (or fibre) width and thickness^{6,31}. A network/mesh of fibres is considered as fully skin imperceptible if it simultaneously fulfils the conditions of: (1) network/mesh opening between fibres greater than $\sim 50\ \mu\text{m}$ (c.f. the sweat gland pore size), but smaller than 1 mm (c.f. the fingertip receptor field); (2) width of individual fibres and thickness of the network/mesh smaller than $\sim 10\ \mu\text{m}$ (such that individual skin cells are mostly exposed through the open fibre network, and the fingerprint ridge features are not compromised). **c,** In situ fibre tethering can be used to couple prefabricated microelectronics and electronic textiles, while supporting on-demand device repair, upgrade, and recycle. Multi-faceted key performance indicators are compared for different methods for fabricating fibre-like building blocks, where the scales of 1-4 are assigned as 4=excellent, 3= very good, 2=acceptable, and 1=needing improvements. The scores are assigned considering literature^{4-6,9,10,13,17,30,31,37,58}, and discussion in Supplementary Note 7 & 8 and Supplementary Table 1.

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Figure 2. Organic bioelectronic fibre fabrication, morphing, and tethering on diverse biological surfaces. **a,** Fibre tethering process, where a fibre is first initiated from an aqueous viscoelastic solution, and then drawn to tether around the target object. An example fibre deposition process is shown on a fingertip. The bioelectronic fibre array transmittance shows a seemingly linear relationship with fibre spacing $\frac{d}{N}$, with the best linear fitting of $T(\%) = 0.045\frac{d}{N} + 89$. **b,** Photos and scanning electron microscope (SEM) images showing the fibre morphing morphologies, for **(i)** A fibre (with a false colour highlight) on a human hair (scale bar = $50\ \mu\text{m}$); **(ii)** Fibres with a red colour dye conform on a dandelion seedhead, and a zoom-in view (scale bars left to right, 1 cm, 1 mm); **(iii)** fibre grids on a Day-3 chicken embryo in a petri dish, and a zoom-in view (with fibres deposited on top of the vitelline membrane covering the yolk; scale bars left to right 5 mm, $500\ \mu\text{m}$). **c,** Fibre surface tethering and wetting on a glass rod (light microscopy time lapse photos), and an orchid flower petal (SEM images, with fibres highlighted with a false blue colour) (scale bars left to right and top to bottom, $500\ \mu\text{m}$, $500\ \mu\text{m}$, $50\ \mu\text{m}$, $10\ \mu\text{m}$, $10\ \mu\text{m}$) **d,** Concepts for fibre patterning, through **(i)** additive (fibre deposition), and **(ii)** subtractive (fibre erasing) processes. (scale bars top to bottom, 5 mm, 5 mm, $500\ \mu\text{m}$)

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Figure 3. Imperceptible on-skin electrodes with tailored formats. **a**, An illustration (top) showing fibres covering the ridges of the fingerprints, where $\frac{N}{d}$ indicates the number of fibres N across a distance d ; and the experimental evidence is provided by the photos (bottom row) showing the complete fibre array on a fingertip, and the zoom-in view of the fibres follow the ridges of the fingerprints (scale bars left to right, 5 mm, 500 μ m). **b**, Contact impedance versus deposition time on the fingertip. **c**, Comparison of ECG signals acquired by fibre and gel electrodes at the same time (signal correlation $P=0.99$). **d**, (i) An array of fibres deposited on the thumb muscle region, where ON/OFF loading on the thumb results in clear on/off EMG signals detected by the fibres (scale bar, 1 cm). (ii), Bar chart to depict variations in absolute EMG amplitude from the thumb muscle region against different loading weight on the thumb (data are presented as mean absolute EMG values \pm standard deviation of EMG measured for around 5 seconds in each case). **e**, Facile repairability of the exposed fibre arrays. The triangular symbol indicates the impedance of the fibre arrays after being deliberately damaged by abrasion, and then new fibres are deposited on-demand to repair as indicated by the circular symbols. **f**, The stability of exposed fibre electrode (exposed bioelectronic fibres on skin) under the conditions of (i) ambient wearing; (ii) mouse clicking; (iii) dry friction wear with a plastic surface (at a surface speed of 4 cm/s under $\sim 40\%$ relative humidity (RH) environments); (iv) simulated ‘wet’ conditions without mechanical disturbance. **g**, Wet friction (at a surface speed of 4 cm/s) of exposed and cellulose-based fibre protected sensing interface. **h**, Rinsing under running water (the sensing interface is protected with cellulose-based fibres and the fibre contact is encapsulated with a cellulose-based film). (ECG scales for **f-g**, horizontal time scale = 1 s, vertical voltage scale = 0.5 mV). (**a-e**, typical results from $N=5$ volunteers, **f-h**, typical results from $N=3$ volunteers, for all experiments with $n>3$ independent experiments performed on each volunteer).

Figure 4. Imperceptible augmentation. **a**, Augmented touch perception via dual-ECG sensing with person-i wearing bioelectronic fibre arrays, and person-ii without. The dual-ECG signal acquired through the fibre array is compared with the reconstructed composite-ECG signal from validation gel electrodes. The red \blacktriangledown and green \blacktriangle symbols indicate the R peaks of person-i and person-ii respectively. **b**, A breathable skin-gated OECT on a fingertip; the OECT displays a response time in the 60 s range. **c**, Dual-modal sensing for augmented perception of mist pulses with acidic, alkaline, and neutral compositions distinguished through colorimetric and electrical readouts. The mist pulse photos shows an example of a neutral mist pulse, and the fibre resistance change was recorded by applying three consecutive neutral mist pulses (normalised resistance change is calculated as $\frac{R^*-R_0}{R_0}$, where R^* is the peak resistance, and R_0 is the initial resistance; the initial resistances of the fibre arrays are in the range of 10 k Ω). (**a-c**, typical results from $N=5$ volunteers, with $n>3$ independent experiments performed on each volunteer).

Figure 5. Adaptable, versatile, and reconfigurable fibre coupling. **a**, Fibre tethering with designable fibre orientations (θ) is demonstrated by statistical analysis of fibre orientations θ

with different fibre patterns, and the patterning accuracies ($P_{\Delta\theta=10^\circ}$) is calculated by taking an 10° -offset (*i.e.*, the width of the binning in the histogram) being an acceptable criterion for misalignment (a horizontal line is used as the 0° baseline for measuring all the fibre orientation angles). **b**, Photos showing the top and profile views of fibre arrays connecting to the contacts of an LED (scale bars, 2 mm). **c**, Distributed bioelectronic fibres to connect with an LED on a plant leaf to warn environmental exposure of ammonia on the plant surfaces (where the dashed lines indicate the boundary of the fibre arrays, scale bars, 5 mm; typical results from independent experiments performed $N>3$ plants). **d**, Concepts for re-usable and re-cyclable components, where the LED and bioelectronic fibres could be separated: the LED reused, and the fibres recycled into a feedstock to create conductive inks for 3D printing (line resistance at $\sim 1 \text{ k}\Omega/\text{mm}$ dependent on filler concentration; scale bars from left to right, and top to bottom, 1 mm, 2 mm, 500 μm). **e**, Concepts for reconfigurable sensing interface. **i**, schematics showing a fibre fabrication and reconfiguration process, where the 'rewrite' process could 'renew' the fibre sensing interface, while retaining similar levels of fibre array resistance between the original R_0 and the 'renewed' R' states; and **ii**, bioelectronic fibre arrays on the surface of a leaf which are written and rewritten to achieve a topological change in the sensing interface (for each fibre array $\frac{N}{d} \sim \frac{60}{1 \text{ mm}}$; scale bars, 5 mm; typical results from independent experiments performed $N>3$ plants). **f**, An array of bioelectronic fibres deposited onto the finger region of an e-textile glove which reduce contact impedance by approximately 2 orders of magnitude, thus enabling biopotential monitoring (scale bar, 200 μm).

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