Mechanically and visually imperceptible sensor sheets integrated with lightweight wireless loggers are employed in ultimate flexible hybrid electronics (FHE) to reduce vital stress/nervousness and monitor natural biosignal responses. The key technologies and applications for conceptual sensor system fabrication are reported, as exemplified by the use of a stretchable sensor sheet completely conforming to an individual’s body surface to realize a low-noise wireless monitoring system (<1 µV) that can be attached to the human forehead for recording electroencephalograms. The above system can discriminate between Alzheimer’s disease and the healthy state, thus offering a rapid in-home brain diagnosis possibility. Moreover, the introduction of metal nanowires to improve the transparency of the biocompatible sensor sheet allows one to wirelessly acquire electrocorticograms of non-human primates and simultaneously offers optogenetic stimulation such as toward-the-brain–machine interface under free movement. Also discussed are effective methods of improving electrical reliability, biocompatibility, miniaturization, etc., for metal nanowire based tracks and exploring the use of an organic amplifier as an important component to realize a flexible active probe with a high signal-to-noise ratio. Overall, ultimate FHE technologies are demonstrated to achieve efficient closed-loop systems for healthcare management, medical diagnostics, and preclinical studies in neuroscience and neuroengineering.

1. Introduction

1.1. Stretchable and Transparent Sensor Sheets

Recently, the use of sensor sheets based on flexible hybrid electronics (FHE), which can operate even under mechanical deformations, has attracted attention for data collection from a wide variety of surfaces, such as the human body, animals, organs, civil structures, and robots.[1–14] For biometric measurements, an extremely soft material is necessary to ensure minimal invasiveness to and inflammation in the living body, such that long-term monitoring is possible to understand neurological mechanisms and diagnose and treat disorders. Silicon semiconductors have ensured high-performance signal processing but such devices exhibit a Young’s modulus between 130 and 170 GPa.[15,16] Considering that the Young’s modulus of a living body is on the order of 1 kPa to 1 MPa,[17–19] it is important to develop a flexible element using an extremely thin polymer, elastomer, or gel specimen that is mechanically equivalent to living tissue.[7,8,13,14,19–25] Thus, stretchable conductors and organic semiconductors remarkably improve the flexibility of the entire sensor sheet (Figure 1a). Another important aspect of such a sensor sheet is its ability to tether an Internet connection when equipped with devices, such as Analog to Digital (A/D) converters, IC chips, or wireless modules, to transfer the collected data in the cloud. In such a case, the sensor sheet can be part of an Internet of things (IoT) system employing closed-loop feedback, allowing technicians to gradually shift from routine on-site inspection to an always-on monitoring system using the sensor sheet under supervised learning. In other words, an Internet-enabling sensor sheet is
a basic requirement to construct a cyber-physical system (CPS) to achieve a streamlined inspection process and solve various problems that hinder applications to, for example, healthcare for humans, animals, civil structures, and robots. Furthermore, a transparent sensor sheet easily allows the visual inspection of an object through devices (Figure 1b–e). Simultaneous physical sensing and optical inspection of the targets can further accelerate the development of the CPS.

A sensor sheet using a stretchable conductor is compatible with the object under inspection even if the object shows surface irregularities and flexibility, which yields high measurement reliability. New materials, such as carbon nanotubes,[20,27,28] graphene,[14,29,30] conductive polymers,[31,32] and metal nanowires,[23,33–35] have been considered as candidates for such a stretchable conductor. Among them, metal nanowires are characterized by higher conductivity than other novel materials, which can be advantageous during wire miniaturization. In addition, networks of metal nanowires have been analyzed as a possible transparent conducting electrode (TCE) that allows light transmission through openings between the nanowires (Figure 1c). Furthermore, a conductive network needs to be maintained even under bending and stretching conditions, which is achieved by strengthening the wire–wire junction.[33,36–41] A FHE using a stretchable TCE can significantly reduce discomfort, invasiveness, and psychological stress for a patient wearing it on their body or having it implanted in an organ. In addition, the epoch-defining method has widened approaches in neuroscience and neuroengineering that can be advantageous during wire miniaturization. In addition, networks of metal nanowires have been analyzed as a possible transparent conducting electrode (TCE) that allows light transmission through openings between the nanowires (Figure 1c).

A key electronic component for such devices comprises a probe introduced on the living body for electrophysiological monitoring. The typical signal amplitude of an electromyogram, electrocardiogram, electrooculogram, and electroencephalogram (EEG), or electrocorticogram (ECoG) ranges approximately from 1 μV to 10 mV.[2,4,5,13,21–23,42–46] An ambitious goal is the ability to probe amplitudes from 1 to 100 μV, produced as a result of brain activity that is extremely small when considering the electromagnetic noise of a random millivolt order in the normal atmosphere. Thus, EEG and ECoG must be acquired by low noise technology before being processed, converted into digital data, and adapted to CPS. Again, the stretchable sensor sheet is essential to be firmly attached to the skin or organ to probe such small signals. This progress report describes the recently developed key technologies that can be employed to implement the aforementioned concept of a mechanically and visually imperceptible sensor system. These technologies include the use of a stretchable probe track, an organic amplifier for an active probe with a high signal-to-noise (S/N) ratio, a metal nanowire based TCE to reduce probe presence, and a lightweight wireless logger to realize an efficient closed-loop system.

Section 1.1 mentioned that it is useful for FHE-based sensor sheets to have stretchability and transparency characteristics for biomonitoring systems. For the entire system, we must integrate a biocompatible flexible sensor sheet with silicon-based rigid components including a wireless module, battery, and A/D converter to obtain an efficient system using CPS. Section 1.2 introduces the methods to provide signal processing capabilities for the flexible sensor sheet. The integration of active components, such as organic transistors and stretchable tracks, is also an indispensable technology. Section 1.3 introduces the metal nanowire based TCE technologies that have stretchability and transparency features. These techniques require comprehension and advancement with the development of mechanical and visually imperceptible sensor sheets. TCE miniaturization technologies are introduced in Section 1.4. Fine fabrication technologies for metal nanowire based TCEs are important to expand the possibilities of a flexible device for the development of multifunctional neural interfaces.

In Section 2, we first demonstrate wireless monitoring systems with flexible sensor sheets. Section 2.1 is an example of EEG monitoring using stretchable tracks (~150%) simply integrated with a silicon-based rigid system.[47] The wireless monitoring system for the EEG had high-quality signals similar to those offered by conventional medical apparatus, which was sufficient to detect Alzheimer’s disease and the status of a healthy person. Section 2.2 describes an example of a system that monitors pulse waves using an FHE system that incorporates active components into the flexible sensor sheet.[48] A developed flexible amplifier was fabricated with organic thin-film transistors to obtain a high S/N ratio and construct a complementary relationship with a silicon semiconductor circuit.

Section 3 reports on the latest progress in TCE technology first introduced in Section 1.3. Techniques that use the metal nanowire based TCE to improve the transparency (~95%), stretchability (~100%), haze (~3%), and electrochemical stability will be discussed in detail. Section 3.1 discusses the synthesis of long and thin Ag nanowires (AgNW), which yielded a high...
transparence and stretchable TCE.\textsuperscript{[39,49,50]} We also remarkably demonstrated a wirelessly obtained ECoG for a neural interface that had a 250 µm width track in a nonhuman primate, i.e., a marmoset under optogenetic stimulation (Section 3.2).\textsuperscript{[44a]} The neural interface containing the AgNW/Au-based track was stable in a rat during a 2 month implantation. All developed technologies for the sensor sheet, which were mechanically and visually imperceptible, show maximized applicability in the diagnosis, treatment, and elucidation of brain disorders.

1.2. Printable Stretchable Tracks and Organic Amplifiers for Flexible Active Probes

A flexible active probe comprising a stretchable track and organic amplifier can provide a high S/N ratio following biological motions. Although organic semiconductors have been reported with excellent durability and resistance to bending and stretching,\textsuperscript{[22,25,51–53]} the ability to ensure excellent properties, such as a suitable voltage threshold, is crucial to establish circuits. Since strains occur in the peripheral joint (−55% strain) and on the skin itself (35–115% strain),\textsuperscript{[54,55]} it is necessary to design a structure that follows the deformations caused by human motion. Therefore, to improve the ductility of the active probe, some attempts have been made to develop a stretchable track that can yield larger deformations as compared with organic circuits.\textsuperscript{[20,21,52,53]}

![Figure 1](image-url) A conceptual image of our flexible hybrid electronics containing a stretchable and transparent track. a) Increased flexibility of the entire sensor sheet on which the stretchable track connects organic components, such as an organic circuit. An Internet-enabling sensor sheet leads to a cyber-physical system. b) A real image of the stretchable transparent conducting electrode (TCE) fabricated with a AgNW connecting LED chip. Optical observations behind the TCE sheet were possible due to high transmittance of visible light. c,d) Schematic of the AgNW-based TCE (c) and its application for electrocorticogram (ECoG) on the cerebral cortex (d). e) A general view of the neural interface including an LED for optogenetically evoked ECoG. f) Adapted with permission.\textsuperscript{[44b]} Copyright 2016, JIEP. e) Adapted with permission.\textsuperscript{[44a]} Copyright 2019, The Authors, published by Wiley-VCH.

Figure 2 shows the resistivity before stretching and maximum strain on the stretchable track as a summary of the conductors investigated thus far. There are two major approaches to achieve a stretchable track: 1) 2D or 3D spring-shaped patterning can be formed by a conventional device manufacturing technology, such as shadow mask evaporation or photolithography\textsuperscript{[2,3,7,13,56,57,63]} and 2) intrinsically stretchable materials or spring-shaped conductors can be fabricated by wet-processing or printing methods.\textsuperscript{[20,23,27–34,39,41,52,58–62,64]} These stretchable tracks have the capability of attaining by 600% strain and are applicable to biological probes,\textsuperscript{[2,3,13,21,23,52]} artificial muscles and skin,\textsuperscript{[27,32,65,66]} solar cells,\textsuperscript{[58,67,68]} and displays.\textsuperscript{[20,63]}

In the context of the first method, the Rogers group has developed a tattoo-type sensor, a sensor for implants with a horseshoe-shaped metal track, and a connecting thin-film silicon.\textsuperscript{[13]} To ensure the horseshoe shape, they used 2D spring-shaped patterning, i.e., nearly a circle, where the track yielded superior stretchability by nearly three-fold as compared with the track without the optimization (Figure 2).\textsuperscript{[3,52]} The Someya group has developed ultrathin (1 µm) transistors and a lightweight (3 g m\textsuperscript{−2}) circuit by evaporating the metal track and an organic semiconductor, which exhibited extreme flexibility, i.e., a bending radii as low as 5 µm.\textsuperscript{[69]} The transistor was less susceptible to distortion when placed on the neutral axis via a 1 µm thick seal.\textsuperscript{[63,68]} The thin film device shaped into a wrinkled structure by prestretching showed stretchability. The Lacour
group also used the prestretching technique to fabricate an Au-based stretchable track on a polydimethylsiloxane (PDMS) substrate.\[56\] The stretchability yielded 100% strain by adjusting the thickness and width of the track to optimize the wrinkled structure. Previous studies have reported this track was extremely flexible and applicable as implantable electronics.\[19\] Conventional device manufacturing technology can produce flexible electronics by incorporating mechanical patterning. The potential disadvantage of this method is friction or interference due to its out-of-plane movement under stretching.

For the second method, intrinsically stretchable materials experience in-plane expansion and contraction, and can be fabricated using printable electronics, resulting in a reduced environmental impact by depositing the required amount of materials at the required locations. In addition, we can achieve a large area of the sensor sheet and high throughput process upon combination with the printing process under a continuous roll-to-roll or sheet-to-sheet process. Cost-effective production can positively contribute to the production of disposable electronics for healthcare applications in the near future. Candidate materials for printable ink and dispersion include not just aforementioned new materials of carbon nanotubes, graphene, conductive polymers, and metal nanowires but metal particles and liquid metals.\[23,33–35,52,61,62,64\] They can be fabricated by simple wet-process techniques, such as coating, drop-casting, and spraying for high-throughput production in printable electronics.

A printable stretchable track comprised of carbon nanotubes, an ionic liquid, and a fluorinated copolymer rubber exhibited a resistivity of $1.8 \times 10^{-2}$ Ω cm that endured up to strains of 118%.\[20\] This stretchable track was patterned by screen printing to produce an organic light emitting diode (OLED) display and active matrix covered with a stiffer. The Someya group further developed a stretchable track using carbon nanotubes, silver flakes, silver nanoparticles, ionic liquid, and a polypyrrolidene diurethane copolymer, obtaining a resistivity of $1.8 \times 10^{-4}$ Ω cm and 140% strain, which was produced by applying a hot-rolling process to reduce the thickness of the track by 5% and ensure uniform filler distribution. A composite of silver flakes and polyurethane rubber exhibited excellent adhesion to different types of substrates. The produced stretchable track exhibited a stretchability at least five times higher due to its elevated affinity for polymer substrates.\[62\] The second method basically does not require sophisticated techniques (i.e., pattern design, prestretching technique, microchannel, etc.) but can further improve stretchability with the techniques.\[27,28,30,12–34,41,58,60,64]\ Overall, stretchable tracks fabricated by a simple manufacturing process does improve the flexibility of the entire device while connecting active elements. Additionally, the printable electronics allow us to obtain versatile and on-demand designs that can advance research.

A challenge in thin-film technologies related to the first method is the fabrication of a passive resistor or capacitor due to size and yield limitations. For example, the thin-film capacitor in the organic amplifier measured nearly 36 $\mu$m\(^2\).\[23\] An organic amplifier must implement alternate components, such as mature chip resistors and capacitors, for miniaturization. This, however, requires a suitable integration technique to apply them to heat-sensitive polymer substrates. A challenge related to the second method is the ability to rapidly and flexibly offer diverse electrode designs. A conventional wearable device, such as a headset that records an EEG, is fixed in terms of design\[45,70\] and can yield a mismatch with the individual's head. Moreover, the headset system can exert pressure on the head (typically less than 13.8 kPa or 2 psi),\[2,4,35,45,70\] which reduces comfort when it is worn. The system showed a noise level of 3–15 $\mu$V or higher\[2,4,35,45\] Thus, a stretchable sensor sheet that can fit a wide variety of surfaces and offers increased softness is necessary to provide long-term use and low-noise technology.

Here, we developed a printable and stretchable sensor sheet that can wirelessly acquire an EEG signal and be worn on the forehead by people ranging in age from infants to elderly adults for extended periods of time (Section 2.1).\[47\] The acquired EEG exhibits the same measurement accuracy as that obtained by a medical apparatus, which can help identify diseases. Moreover, the organic amplifiers successfully integrated within the chip-type resistors and capacitors on the flexible polyelephene naphthalate (PEN) were fabricated using FHE, with which pulse waves were amplified and wirelessly transmitted to a PC (Section 2.2).\[60\] This technique allows the fabrication of a flexible active probe that uses stretchable tracks and organic semiconductors, which should to act as a complementary system between the electrode probing biosignals and a silicon-based chip processing signal to reduce background noise.
exhibits a light transmittance of 86% and sheet resistance of indium-tin-oxide (ITO)-based TCE. The AgNW-based TCE bending radii as low as 4 mm, not exhibited by the conventional optical and electromagnetic properties different from those of bulk, catalytic species due to their large specific surface area.\textsuperscript{[71–75]} To elicit these properties, we must use shape control of the nanomaterial. In 1989, the Figlarz group proposed the “polyl method” as a simple chemical synthesis to reduce the ions in polyhydric alcohols.\textsuperscript{[76]} Using the polyl method, it is possible to control the shape of the metal nanomaterials, such as spherical nanoparticles, nanowires, nanotubes, nanoprisms, and nanoplates.\textsuperscript{[77–79]} Overall, high metal nanowire yields have been achieved through chemical synthesis, which can be applied to flexible wiring materials and optical devices in electronics.\textsuperscript{[80–82]}

Lee et al. (Stanford University) developed the Ag nanowire (AgNW)-based TCE in 2008.\textsuperscript{[82]} This electrode offers flexibility, bending radii as low as 4 mm, not exhibited by the conventional indium-tin-oxide (ITO)-based TCE. The AgNW-based TCE exhibits a light transmittance of 86% and sheet resistance of 16 $\Omega$ sq.\textsuperscript{−1}, equivalent to that of the ITO-based TCE. Since then, several studies have attempted to improve the characteristics of the AgNW-based TCE, such as its flexibility, conductivity, and transparency. Meanwhile, the TCE has been applied in organic solar cells, organic lighting, and touch panels.\textsuperscript{[80,83–86,88–85]}

The TCE can be produced by introducing synthesized AgNW on a transparent substrate (Figure 1). For example, AgNWs were chemically synthesized by reducing Ag nitrate in an ethylene glycol (EG) solvent, which contained dissolved polyvinyl pyrrolidone (PVP) and chloride ions acting as a capping and shaping agent, respectively.\textsuperscript{[49,50,81]} The synthesized AgNW consisted of five single crystals with triangular cross-sections, growing in the (110) direction during synthesis.\textsuperscript{[80]} In fact, the cross section of the silver nanowire was characterized by a pentagonal cross section formed by five single crystals. After synthesis, the reaction solution is typically centrifuged and replaced with an alcohol solvent, such as ethanol. When the AgNW dispersion is deposited and dried on a transparent substrate to increased pressure, which is beneficial for simple pressing techniques can damage delicate substrates or devices.\textsuperscript{[33,36–38,40,41,68,80,85]} The junctions are mechanically and electrically improved using these methods. As a result, nanowire networks maintain their conductivity with exposure to deformation. However, thermal annealing (often over 150 °C) is time consuming (i.e., taking minutes to hours) and excludes the use of many heat-sensitive materials while pressing techniques can damage delicate substrates or devices. The second challenge is that Ag-based conductors tend to electrically break during short-term use, which is attributed to atmospheric corrosion, i.e., the presence of sulfides, and atomic migration, i.e., via joule heating.\textsuperscript{[95–102]} This phenomenon is accelerated in the presence of water and moisture. Recently, an insulation layer between AgNW and air was introduced in the form of a photore sist, polymer layer, and reduced graphene oxide flakes.\textsuperscript{[103–105]} However, these barriers have intrinsically low electrical conductivity.

Here, we propose a method to improve the properties of the AgNW-based TCE (Section 3.1), which involved synthesis to obtain long and thin AgNWs for high transmittance and low haze.\textsuperscript{[49,50]} annealing by high-intensity pulsed light (HIPL) to enhance the mechanical stability,\textsuperscript{[39]} and treatments via Au plating to enhance the electrical reliability. The fabrication involved a low-temperature process that did not subject the substrate to increased pressure, which is beneficial for simple and effective assembly in the FHE that integrates organic components.

### 1.3. Metal-Nanowire-Based TCE

Possibly applicable in electronics, metallic nanomaterials have optical and electromagnetic properties different from those of bulk, catalytic species due to their large specific surface area.\textsuperscript{[71–75]} To elicit these properties, we must use shape control of the nanomaterial. In 1989, the Figlarz group proposed the “polyl method” as a simple chemical synthesis to reduce the ions in polyhydric alcohols.\textsuperscript{[76]} Using the polyl method, it is possible to control the shape of the metal nanomaterials, such as spherical nanoparticles, nanowires, nanotubes, nanoprisms, and nanoplates.\textsuperscript{[77–79]} Overall, high metal nanowire yields have been achieved through chemical synthesis, which can be applied to flexible wiring materials and optical devices in electronics.\textsuperscript{[80–82]}

Table 1. TCE performance fabricated with various materials.\textsuperscript{[36,49,80,89,93,94,129–133]} Adapted from ref. [156]. Copyright 2019, Wiley-VCH.

| Parameter          | Typical AgNW | CNT & Graphene | ITO   | LT-AgNW |
|--------------------|--------------|----------------|-------|---------|
| Optical transmittance | 90%          | 90%            | 85%   | 94–97%  |
| Sheet resistance    | 20–100 $\Omega$ sq.\textsuperscript{−1} | >100 $\Omega$ sq.\textsuperscript{−1} | 5 $\Omega$ sq.\textsuperscript{−1} | 24–109 $\Omega$ sq.\textsuperscript{−1} |
| Haze               | 5–15%        | <1%            | 1–3%  | 1.6–3.4% |

Haze 5–15%
Table 2. Mechanical flexibility under bending tests.

| Material | Bending radii/ΔR R \(^{1,4}\) | Strain | Substrate/thickness [μm] | Key factor | Process/temperature/heating time | Ref. |
|----------|-----------------------------|--------|------------------------|------------|----------------------------------|------|
| ITO      | 0.5 mm/O.L. for a 20 nm thickness 4 mm/O.L. for a 100 nm thickness | 0.4%   | PI/40                  | –          | Magnetron sputtering/N.D./N.D.  | [111]|
| PEDOT/PSS| 10 μm/N.D.                  | –      | PET/1.4                | Prestretched with elastomer | Drying/120 °C/30 min | [63,67]|
| CNT      | 1 mm/≈1.5%                  | 4%     | PET/188                | Shear stress by brush printing | Drying/150 °C/5 min | [112]|
| Graphene | 15 μm/20%                   | –      | PI/10                  | Entangled structure | CVD/1050 °C/1 h Transfering/\( T_{\text{room}}/N.D. \) | [113]|
|          | 1.5 mm/>20%                 | 1.1%   | PET/>5                 | –          | CVD/N.D./N.D.→Transfering/N.D./N.D. | [114]|
|          | 0.5 mm/>5%                  | –      | PDMS/2000              | Wavy structure | CVD/1000 °C/1 h hPDMS curing/80 °C/1 h | [115]|
| AgNWs    | 30 μm/3.5%                  | 13%    | PI/7–8                 | Embedded into thin substrate | PI curing/150 °C/1 h | [116]|
|          | ≈1 mm/>1%                   | 0.6%   | PI/5–6                 | Embedded into thin substrate | Drying/100 °C/10 min–PI curing/180 °C/10 min | [117]|
|          | 200 μm/25%                  | 0.25%  | NOA 63⁴/100           | Embedded into thin substrate | Drying/100 °C/5 min–UV irradiation/\( T_{\text{room}}/10 \) min | [118]|
| AgNFs    | 0.5 mm/N.D.                 | –      | PET/–                  | Low junction resistances | Sintering/280 °C/2 h | [119]|
|          | 1 mm/N.D.                   | –      | PET/–                  | Low junction resistances | Sintering/250 °C/2 h | [120]|

\(^{1}\)\((R_{\text{meas}} - R_{0})/R_{0}\). \(^{2}\)R_{\text{meas}}; resistance measured while being bent, \( R_{0} \); initial resistance; \(^{3}\)A commercial ultraviolet curable polymer from Norland Optical Adhesive (NOA) 63.

1.4. TCE Miniaturization

Miniaturization of the stretchable TCE can extend its application to devices that require high resolution, such as neural interfaces, biosensors, and field effect transistors. Microelectrodes that can monitor brain activity, such as the acquisition of an ECoG, must be designed to target a single-unit cell or group of neurons in rodents, nonhuman primates, and humans. As a result, the contact impedance to devices that require high resolution, such as neural interfaces, biosensors, and field effect transistors.

Graphene and metal nanowires have been confirmed in novel materials made of conductive polymers, carbon nanotubes, graphene, and metal nanowires. However, brittleness of the ITO, which breaks down at 0.4% strain, renders fabrication on an elastomer substrate difficult. As listed in Table 2, higher flexibility than ITO have been achieved by an additive method, e.g., a wet printing process, such that the conductive network is not ruined.

The AgNW-based TCE, for example, has a random mesh structure constructed in a similar manner to a carbon nanotube-based TCE. When the silver nanowire electrode forms a network, the conductivity path of the wire may reduce the conductivity performance of the wire. Meanwhile, a thicker AgNW network reduces the optical transmittance. Thus, the miniaturization of the AgNW-based TCE by an additive method is optimal, with circuit fabrication only at necessary locations using an optimal amount of ink to securely form the random network inside the track. Table 3 lists the resolution, conductivity, and process temperature for each novel material. Again, the AgNW-based TCE was characterized by high performance on a polymer substrate even after fine patterning. There are several studies that investigate printing a AgNW-based track onto a polymer substrate via the subtractive method, reductions in the conductive path of the wire may reduce the conductive performance of the wire. Thus, the miniaturization of the AgNW-based TCE by an additive method is optimal, with circuit fabrication only at necessary locations using an optimal amount of ink to securely form the random network inside the track. Table 3 lists the resolution, conductivity, and process temperature for each novel material. Again, the AgNW-based TCE was characterized by high performance on a polymer substrate even after fine patterning. There are several studies that investigate printing a AgNW-based track onto a polymer substrate via the subtractive method.
neural interface because the resolution it offered was sufficient to record the neural activity of a cell group. Finally, we used the stretchable and transparent TCE combined with the Au-plated AgNW to wirelessly record an ECoG for long-term implantation (Section 3.2).\[44]\] Additionally, the wireless monitoring system, implemented with LED illumination, recorded a marmoset’s ECoG during an optogenetic stimulation.

2. Wireless Monitoring System

2.1. Electroencephalogram Monitoring System with a Stretchable Track

A wireless monitoring system that can obtain an EEG when placed on the forehead yields a new method to perform a simple brain diagnosis that can be part of a CPS, which does not result in discomfort due to the use of a stretchable sensor sheet (Figure 3).[47] The stretchable sensor sheet (weight = 0.5 g, thickness = 80 µm, stretchability ≤ 150%) in the monitoring system was fabricated by screen-printing a Ag particle—containing paste onto the elastomer substrate that had a moisture permeability of up to 2700 g m⁻² day⁻¹ (25 µm thickness at 40 °C and 90% humidity), which can help avoid bacterial growth even in a wet state. In addition, its total thickness of less than 0.1 mm also reduces discomfort compared with the use of commercialized electrodes with a thickness of ≈1–10 mm. Multichannel and low-noise analog front ends (0.25–1 µV at 2–30 Hz) were implemented in the wireless module (weight of 12 g, 6 mm thickness) to attain expandability in the wireless monitoring system, which includes PC communication to display the EEG and acquiring rapid Fourier transform (FFT) results in real time. Brain waves potentially provide a variety of information, such as the possibility of epilepsy, Alzheimer’s disease, impaired consciousness, sleep quality, and the degree of stress or relaxation.[45] Thus, using our wireless monitoring system, we have developed a simple and agreeable brain diagnosis method to monitor a person’s health, which is as easy as measuring body temperature using a thermometer.

We compared the wireless monitoring system with commonly used medical apparatus. The system platform has an 8-channel low noise A/D converter, lithium-ion battery, and Bluetooth low energy (BLE) wireless communication modules on a flexible board. Its total size is 3 cm × 9 cm × 6 mm and weighs only 12 g. EEG measurements with both our system and the medical apparatus (Neurofax; Nippon Opto-electronics, Japan) were conducted by placing the electrodes at a distance of less than 1 cm from the Fp1 (left-Frontal Pole). The subject was kept in an anechoic room with controllable lighting during measurements sampled at 250 and 500 Hz using our system and the conventional apparatus, respectively. As shown in Figure 3b, our system confirmed high consistency based on the results that showed a value of more than 0.98 for the coefficient of correlation between the waveforms. The EEG in Figure 3b was observed in a closed-eye state. The characterized resistance of the stretchable track was less than 1.5 kΩ. We can rationalize such a high resistance considering a skin-contact impedance of 2–55 kΩ (measured at 10 Hz). Subsequently, we proposed a method to diagnose Alzheimer’s disease (AD) using our system. Figure 3c shows the relationship between power spectrums at 5–8 Hz and 19–21 Hz for brain waves in the monitored Fp1.[146] Based on these results, we obtained a classification error of 12.5% (2 out of 16 subjects). This indicates that a simple diagnosis is possible based on the spectral information. Although higher accuracy is a future goal, we have demonstrated that techniques to integrate elastic materials with printing technology are successful and possible to implement mass production for rapid in-house brain scans to confirm if a more thorough medical checkup is necessary.

Table 3. Fabrication parameters for the printed TCE and its performance with respect to miniaturization.

| Material   | Fabrication method | Widths/spacings | Thickness | Visible transmittance | Conductivity or sheet resistance | Annealing conditions | Ref. |
|------------|--------------------|-----------------|-----------|-----------------------|----------------------------------|---------------------|------|
| PEDOT/PSS  | Hydrophilic/hydrophobic pattern | 50/≈150 µm | 80–350 nm | 89–96% | 0.6–1.8 S cm⁻¹ (3–4 kΩ sq⁻¹) | 140 °C, 30 s | [124] |
|            | Ink jet            | 65 µm | 301 nm | 80% | 5.0 × 10⁻² S cm⁻¹ (66 Ω sq⁻¹) | 80 °C, 15 min | [125] |
|            | Transfer           | 95/100 nm | 100 nm | – | 7.6 × 10⁻³ S cm⁻¹ | – | [126] |
| CNT        | Ink jet            | 70/≈ µm | – | – | 4.0 × 10⁵–3.0 × 10⁶ Ω sq⁻¹ | – | [127] |
| Graphene   | Ink jet            | 60/≈ µm | 14–150 nm | – | ≈2.5 × 10⁴ S cm⁻¹ | 250 °C, 30 min | [129] |
|            | Gravure            | 25/25 µm | 15–30 nm | – | 1.0 × 10⁵ S cm⁻¹ | 250 °C, 30 min | [130] |
|            | Screen             | 40/30 µm | 0.1–2 µm | – | ≈1.9 × 10⁵ S cm⁻¹ | 300 °C, 30 min | [131] |
| AgNWs      | Gravure            | 450/100 µm | – | ≈90% including spaces | 14 Ω sq⁻¹ | 90 °C, 10 min | [133] |
|            | Ink jet            | 50/70 µm | 0.5–2 µm | ≈85% | 35–100 Ω sq⁻¹ | 100 °C, 30 min | [134] |
|            | Screen             | 50/50 µm | 0.2–1 µm | – | ≈4.7 × 10⁵ S cm⁻¹ (≤ 1.1 Ω sq⁻¹) | 150 °C, 15 min | [135] |
|            | Hydrophilic/hydrophobic pattern | 50/50 µm | – | ≈70% | 537 Ω sq⁻¹ | 120 °C, 60 min | [136] |
2.2. Pulse Wave Monitoring System with Organic Amplifier

An organic thin-film transistor (OTFT) was fabricated with shadow mask evaporation on a PEN film (50 µm thickness) and connected using assembled mature chips of resistors and capacitors to produce an amplifier, which can monitor an amplified pulse wave using wireless communication (Figure 4). The OTFT comprised an Al gate, AlOx and Parylene gate insulator, dianthra[2,3-b:2',3'-f][thieno[3,2-b]thiophene (DNTT) semiconductor, Au source, and Au drain (Figure 4a–c). Subsequently, a Ag-based conductive adhesive was deposited and cured at 120 °C for 60 min. This process did not affect the properties of the OTFT. The conductive adhesive had a resistivity of 1.0 × 10⁻² Ω cm, and was mounted with a 0 Ω chip maintaining a 15 µm-space from the substrate that was completely intact (Figure 4d). The electrical resistance was maintained and did not change after 10 bending cycles with a curvature radius of 2.5 cm. This was attributed to durable bonding, a shear strength of 71–92 gf, and elastic modulus for the conductive adhesive itself of 5.1 GPa. Finally, a chip resistor and chip capacitor (1 Ω Mµ) were mounted onto the OTFT circuit to fabricate an amplifier, whose biosignal amplification was monitored using our wireless module (Figure 4e). A flexible piezoelectric film was laid on the wrist to detect vibrations due to vascular constriction. The output voltage of the piezoelectric film was amplified 30-fold. Thus, the active probe provided a high S/N ratio to reduce background noise. As a proof of concept, we were then able to confirm that we had successfully developed a stretchable track and flexible organic amplifier that transferred the signal to the wireless module described in Section 2.1 and 2.2.

3. Metal Nanowire Based TCE with High Performance and Reliability

3.1. Long and Thin AgNW for High Transparency and Stretchability

We synthesized long and thin AgNWs (LT-AgNWs) at low temperatures (110–130 °C) without strong agitation using the polyl method (Figure 5). The diameters of these AgNWs were 60–100 nm and lengths were 20–100 µm, which is three times longer than AgNWs normally synthesized at 150 °C under high agitation at 700 rpm. By reducing the mechanical agitation to 0–60 rpm, we were able to grow long AgNWs using the simple one-pot polyl process (Figure 5c). Additionally, the low temperature and stationary solution provided a sufficient growth time for the seeds. The diameter appears smaller than those obtained using previously mentioned methods due to uniform nucleation growth.

Subsequently, we evaluated the electrical and optical performance of the LT-AgNW-based TCE prepared by drop-coating an ethanolic dispersion on a glass substrate. The TCE formed with AgNWs had an aspect ratio of 484, nearly three times higher than that of a typical AgNW-based TCE. Additionally, it could attain a haze of 3% or less after annealing, which is equivalent to uniform nucleation growth.

The LT-AgNW-based TCE had a sheet resistance six times lower than that present in the wire–wire junction. In fact, the LT-AgNW-based TCE yielded a sheet resistance as low as 19 Ω sq.−¹ and a parallel light transmittance of 80% even without annealing (Figure 5e). The LT-AgNW-based TCE had a sheet resistance six times lower than that of a typical AgNW-based TCE. Additionally, it could attain a haze of 3% or less after annealing, which is equivalent...
to that of the transparent and conductive ITO film (Figure 5f and Table 1). Along with haze of less than 3% and transmittance of 94–97%, the sheet resistance was also maintained at a low value of 24–109 Ω sq.−1. On the other hand, the typical TCE composed of carbon nanotubes or graphene generally exhibit a sheet resistance of 100 Ω sq.−1 or more, haze below 1%, and transmittance of 90%. Against the single-layer graphene-based TCE (125 Ω sq.−1 at 97%), the LT-AgNW-based TCE was characterized by a higher conductivity and light transmittance at a comparable haze value that exceeded those of the ITO.

To investigate the stretchability of the LT-AgNW-based TCE, the TCE was fabricated on an elastomer substrate. Figure 6a shows the fabrication process using spray coating with ethanol-based dispersion on a transparent polyurethane substrate (100 µm thickness, MG; Takeda Sangyo Co., Ltd.), followed by annealing using HIPL (PulseForge 3300; Novacentrix). The HIPL generated localized heat on the metallic nanostructure and induced nanowelding between nanowires, which are both due to surface plasmon resonance. Although the capping agent covered the AgNWs and there was a gap of several tens of nanometers at the wire–wire junction, light energy with a pulse duration of 50 µs effectively elevated the temperature, estimated at over 700 °C. Thus, the sheet resistance of the LT-AgNW-based TCE decreased dramatically, i.e., from 11 to 2 Ω sq.−1 at a transmittance of 50% and from nearly 100 to 9 Ω sq.−1 at a transmittance of 80%. Additionally, the polyurethane substrate softened near the AgNW during HIPL, with an estimated depth less than 10 µm from the top surface, but remained at room temperature on the bottom side due to low thermal conductivity (0.03 W mK−1). In fact, fully or partially embedded AgNWs on the surface of the substrate were observed (Figure 6b,c). This phenomenon was confirmed by a simple experiment involving a tape test. Adhesion between...
the AgNWs and transparent elastomer substrate was strong and exhibited limited exfoliation during the tape test although the simply coated AgNW not subjected to HIPL completely peeled off. Since the LT-AgNW-based TCE treated with HIPL had a mechanically strong structure, contact between the wires was not easily loosened during expansion and contraction. Figure 6d,e shows the electrical resistance during the stretching test. The LT-AgNW-based TCE treated with HIPL maintained a low resistance under a one-time stretching test of up to 100% strain and during 1000 cycles of 20% strain. In contrast, rapidly increased electrical resistances were detected in the cyclic test for the simply coated AgNWs and the one-time stretching test for the short AgNWs (Figure 6d,e). Therefore, the combination of using LT-AgNWs and a mechanically strong structure is the most effective method to obtain a highly stretchable TCE.

Considering that a Ag-based conductor increased its resistance due to long-term exposure in air, as described in Section 1.3, improvements to its electrical reliability are necessary for their use in electronic devices. For this purpose, we introduced electroless Au plating on LT-AgNWs (AgNW/Au). Figure 7a shows a cross-sectional image of the AgNW/Au obtained via electron microscopy. For the AgNW/Au, we observed a uniform Au layer that was several nanometers thick on the AgNW surface and localized depositions of Au at the wire–wire junction. The latter effect from “Au nanobonding” apparently contributed to the enhanced stretchability compared to that of the pristine AgNW (Figures 6d and 7b), which did not carry over to the effects of HIPL. Importantly, the AgNW/Au had a high electrical durability during the tests when the applied constant current values were 1, 10, 20, 30, and 40 mA on the AgNW/Au track in the presence of a water drop (Figure 7c). These tests impose severe acceleration due to electrochemical migration. The AgNW/Au track fully withstood an electrical load of $1.4 \times 10^5$ J while the pristine AgNW track broke down at an electrical load that was...
20 times lower than that applied to the AgNW/Au track. The AgNW/Au exhibited a fully uniform core–shell structure that contributed to restraining corrosion and atomic migration. Thus, we conclude that the Au plating on the LT-AgNW-based TCE resulted in high electrochemical and mechanical stability, which can contribute to successful biometric measurements using the transparent and stretchable TCE.

3.2. Wireless Monitoring Using an Optogenetic Neural Interface for Electrocorticogram Monitoring

We developed a wireless module implemented with 64-channel and 2-channel LED drivers for simultaneous optogenetic manipulation and potentiometric monitoring (Figure 8a). The main board (75 mm × 40 mm × 12 mm and mass of 33.53 g) and A/D converter board (25 mm × 20 mm × 2.5 mm and mass of 0.98 g) were of suitable sizes for animals to wear or be implanted with. Figure 8b compares the ECoG recorded using our system and that obtained with a conventional electrophysiological system (Cerebus; Blackrock Microsystems LLC, Salt Lake City, UT, USA). The two microelectrodes were placed near each other, at a distance of 200 µm, on the somatosensory cortex of a rat under anesthesia. The ECoG recorded with our system (at 1 kHz sampling) was comparable to that of the conventional system (at 2 kHz sampling). Remarkably, our wireless module yielded low noise (less than 1 µV) while being small, lightweight, and applicable for a connection to a neural interface.

Recently, a transparent and flexible neural interface was developed with a AgNW/Au-based track, which can be integrated with an LED probe that has a 50 µm thickness and 500 µm width (Figure 8c,d). A AgNW/Au-based track, with a width of 0.25 mm, was patterned by laser etching, exhibiting an electrical durability that was 20-fold higher than that of the...
pristine AgNWs. In fact, the neural interface, which was built with an inner track and outer hydrogel–gel-based microelectrode on an ultrathin Parylene-C substrate, had a low impedance of 1.1–3.2 $\Omega \text{cm}^2$ maintained in a saline solution over a 5 month period. Additionally, biocompatibility was demonstrated based on immunohistochemical tests conducted on a rat for a 5 month implantation. Thick granulation tissue did not show sufficient grow as is typically the case due to the coating of an antithrombogenic polymer. Thus, somatosensory-evoked potentials (SEPs), related to the integrity of neurotransmission, were observed after the 2 month implantation in the rat (Figure 8e).

Optogenetic stimulation and ECoG monitoring in a marmoset’s cerebral cortex was performed with a neural interface, which showed excellent performance when connected to our wireless module. Optogenetics is widely used as a technique to activate or inhibit targeted neurons via light.[3,7,42–44a] Viruses (AAV5-Syn-Chronos-GFP; Vector Core, University of North Carolina, Chapel Hill, NC, USA) were infused into the brain at the motor cortex. When a blue LED (wavelength: 465 nm) was illuminated on the viruses at the injected point with a pulse duration of 1 ms and illuminance $< 4.6$ mW mm$^{-2}$, an increased response intensity was observed (Figure 8f). On the other hand, intensity decreased when the LED probe was placed at a distance from the injected area. The neural interface, which can be tuned with a total transmittance within a range of 69–83%, can expand the applicability of our system to reduce blind spots and ensure light penetration. Thus, our wireless monitoring system can successfully perform long-term ECoG acquisition and remote optogenetics.

4. Conclusions
We have discussed the challenges concerning the use of sensor sheets, which are required for their stretchability, transparency, and long-term stability for their use in biological monitoring systems. We proposed techniques using metal nanowires, OTFT, and a wireless module to fabricate devices that can acquire an EEG, ECoG, and monitor pulse waves. We can conclude with the following the following summary of the findings.

1) **AgNW-based TCE**: We proposed methods to improve the properties of the AgNW-based TCE. Synthesis at a low temperature, i.e., 110 °C, and under low agitation, i.e., less than 60 rpm, elongated the AgNWs (20–100 µm) while maintaining a thin diameter (60–100 nm). The LT-AgNW-based TCE proposed is characterized by a low haze (<3%) and high...
transparency (<97%). HIPL and Au plating treatments enhanced the stretchability (when exposed to 100% strain) and electrical reliability. All processes were performed at low temperatures and did not involve the exertion of pressure on the substrate, which suggests the ability to simply and effectively assemble the FHE that integrates organic components.
2) **Wireless module for the EEG and ECoG:** A lightweight and small wireless module was developed with 8ch for the EEG and 64ch for the ECoG. Measurement accuracy was at a level similar to that offered by medical apparatus in both cases during real measurements conducted on the human forehead and a rodent’s somatosensory cortex. A 2ch driver for LED was implemented in the wireless module for the ECoG, which is useful for optogenetic manipulation.

3) **Sensor sheet for the EEG:** We developed a printable and stretchable (<150%) sensor sheet (opaque) that wirelessly acquires an EEG for the first time, which can identify Alzheimer’s disease and the healthy status of a person. Moreover, a noise level of 0.25–1 μV at 2–30 Hz was observed during the EEG measurements. Thus, an imperceptible sensor sheet fitted onto an individual’s forehead can offer rapid in-home brain diagnosis and in healthcare sectors.

4) **Neural ECoG interface:** The neural interface was fabricated with a AgNW/Au-based track (250 μm width) patterned by a subtractive method and a hydrogel-coated microelectrode. The system, with an antithrombogenic polymer, was stable in a rat during a 2 month implantation. Additionally, we developed a neural interface that wirelessly monitored an optogenetically evoked ECoG in a marmoset under illumination by an implemented LED. The simultaneous stimulation and monitoring can contribute to a multifaceted approach for diagnosis and treatment in neurological studies.

5) **OTFT:** A DNTT-based OTFT was implemented with a chip resistor and capacitor on a flexible PEN substrate to fabricate an organic amplifier using FHE. The amplifier achieved 30-fold amplification of the pulse waves, where a wireless module transmitted the signals to a PC.

The developed sensor sheet fabrication technology can be manipulated using stretchable tracks and organic semiconductors to achieve biocompatibility. We expect that the sensor sheet can act as a transducer yielding a high S/N ratio and feature reduced background noise, which should allow one to build a complementary system between a flexible active probe and silicon-based processor to wirelessly transmit data. Thus, the developed sensor sheet maximizes its potential in the CPS for applications in the fields of medical care, civil structure engineering, robotics, sport, and entertainment.

**Conflict of Interest**

The authors declare no conflict of interest.

**Keywords**
electrophysiology, flexible electronics, metal nanowires, transducers, wireless recording

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