Kent Academic Repository

Wang, Zixin, Mahdi, Saleh, Alwatter, Aula A., Alfredsson, Maria, Horne, Robert, Chee Siang, Ang, Condell, Joan, Flowers, Emily, Matcham, Faith, Robinson, Tony and others (2025) ***Evaluation of Carbon-Based Interconnects for Digital Signaling in Printed Flexible Electronics on Sustainable Substrates.*** **IEEE Journal on Flexible Electronics .**

Downloaded from

https://kar.kent.ac.uk/115148/ The University of Kent's Academic Repository KAR

The version of record is available from

https://doi.org/10.1109/JFLEX.2025.3562530

This document version

Publisher pdf

DOI for this version

Licence for this version

UNSPECIFIED

Additional information

Versions of research works

Versions of Record

If this version is the version of record, it is the same as the published version available on the publisher's web site. Cite as the published version.

Author Accepted Manuscripts

If this document is identified as the Author Accepted Manuscript it is the version after peer review but before type setting, copy editing or publisher branding. Cite as Surname, Initial. (Year) 'Title of article'. To be published in ***Title of Journal*** , Volume and issue numbers [peer-reviewed accepted version]. Available at: DOI or URL (Accessed: date).

Enquiries

If you have questions about this document contact ResearchSupport@kent.ac.uk. Please include the URL of the record in KAR. If you believe that your, or a third party's rights have been compromised through this document please see our Take Down policy (available from https://www.kent.ac.uk/guides/kar-the-kent-academic-repository#policies).

Evaluation of Carbon-Based Interconnects for Digital Signaling in Printed Flexible Electronics on Sustainable Substrates

Zixin Wang, Mahdi Saleh, *Member, IEEE*, Aula Alwattar, Maria Alfredsson, Robert J. Horne, Chee S. Ang, Joan Condell, Emily Flowers, Faith Matcham, Tony Robinson, John Rooksby, Sian Saha, Louise Rose, John C. Batchelor, *Senior Member, IEEE*, and Alexander J. Casson, *Senior Member, IEEE*

***Abstract*—Printed electronics using flexible substrates are an emerging area, allowing next-generation electronics to conform and flex with different surfaces, from human skin to clothing. In the hybrid integration or sea-of-rigids, approach, conventional microchips are mounted onto (generally) plastic substrates such as polyethylene naphthalate (PEN), with (typically) printed silver tracks for interconnections between components. An ongoing research direction is to replace plastic substrates with biodegradable substrates and to replace silver tracks with nonheavy metal-based tracks. While the substrates and tracks form only part of an overall system, replacing them is a step toward increased sustainability and helps to meet net-zero goals for printed electronic systems. Previously, several papers have investigated printed carbon tracks for low-frequency analog sensing applications. This article explores the feasibility of using printed carbon tracks on biodegradable substrates for high-frequency applications such as digital signaling over a serial-peripheral interface (SPI). We investigate the printability, thermal stability, and electrical conductivity of carbon ink screen-printed onto six commercially available sustainable and flexible substrates. Our results demonstrate that multilayer screen printing substantially reduced the electrical resistance of carbon tracks, enabling SPI communication at frequencies up to 16 MHz with three layers of carbon ink. A Natureflex substrate provided the best balance of printability, thermal stability, and electrical performance. Substrates such as greaseproof paper and ClearFilm PU showed potential for flexible electronics, but require further optimization. This study provides valuable insights into selecting and optimizing biodegradable substrates for high-frequency digital systems, supporting the move toward more sustainable printed electronics.**

***Index Terms*—Digital communication, flexible electronics, impedance, screen printing, sustainable electronics.**

I. Introduction

FLEXIBLE electronics have received substantial attention in the research literature [1], with applications ranging from wearable devices [2] to resistive, capacitive, and electrochemical sensors [3]. Printing-based manufacturing of flexible electronics is a widely used approach [4] as it allows thin, flexible, and sometimes stretchable, electronics suitable for challenging applications that are not possible with traditional printed circuit boards (PCBs) (such as FR-4) while being compatible with scaled-up, industrial processes such as roll-to-roll printing [5].

Recently, there has been a significant focus on enhancing sustainability in electronics and minimizing electronic waste. Electronic waste is one of the largest growing waste streams worldwide, motivating substantial research into biodegradable electronics [6]. These Biodegradable electronics can offer a more sustainable approach compared to traditional substrates such as FR-4, and new use cases for novel flexible electronic systems [7]. For example, in healthcare applications, they may allow using invasive devices that can be surgically placed inside the body and then naturally degrade when reaching their end of life, eliminating the need for removal surgery [8]. Or, in noninvasive monitoring, they may allow items that can break down at the end of use [9].

Another key aspect supporting the move to printed electronics is sustainability [10], [11]. Local additive manufacturing techniques reduce the environmental impact of traditional manufacturing processes, and potentially the need for long-distance transportation. While the end-of-life options for printed electronics have been considered for some time, the 2021 flexible and printed electronics roadmap [12] gives minimal attention to this important topic. More recent roadmaps

Received 23 February 2025; accepted 6 April 2025. Date of publication 5 May 2025; date of current version 8 July 2025. This work was supported in part by U.K. Engineering the Physical Science Research Council under Grant EP/S020179/1 and Grant EP/X000788/1; and in part by the Henry Royce Institute for Advanced Materials through EPSRC under Grant EP/R00661X/1, Grant EP/P025021/1, Grant EP/P025498/1, Grant EP/S020179/1, and Grant EP/X000788/1. *(Corresponding author: Alexander J. Casson.)*

Zixin Wang, Mahdi Saleh, and Alexander J. Casson are with the Henry Royce Institute for Advanced Materials, M13 9PL Manchester, U.K., and also with the Department of Electrical and Electronic Engineering, School of Engineering, The University of Manchester, M13 9PL Manchester, U.K. (e-mail: alex.casson@manchester.ac.uk).

Aula Alwattar is with the Department of Chemistry, School of Natural Sciences, The University of Manchester, M13 9PL Manchester, U.K.

Maria Alfredsson is with the School of Chemistry and Forensic Science, University of Kent, CT2 7NZ Canterbury, U.K.

Robert J. Horne and John C. Batchelor are with the School of Engineering, University of Kent, CT2 7NZ Canterbury, U.K.

Chee S. Ang is with the School of Computing, University of Kent, CT2 7NZ Canterbury, U.K.

Joan Condell and Tony Robinson are with the School of Computing, Engineering and Intelligent Systems, Ulster University, BT48 7JL Derry, U.K.

Emily Flowers, Sian Saha, and Louise Rose are with the Division of Digital Health and Applied Technology Assessment, King's College London, WC2R 2LS London, U.K.

Faith Matcham is with the School of Psychology, University of Sussex, BN1 9RH Brighton, U.K.

John Rooksby is with the Department of Computer and Information Sciences, Northumbria University, NE1 8ST Newcastle upon Tyne, U.K.

Digital Object Identifier 10.1109/JFLEX.2025.3562530

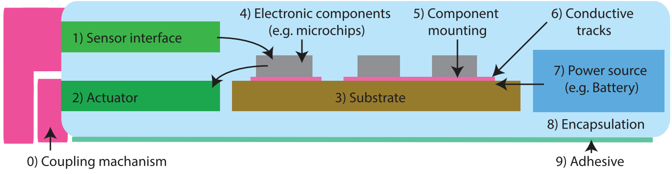

Fig. 1. Cross section of a low-power sensor node device highlighting the many system blocks that need to be considered for sustainability. This work considers the substrate primarily.

on flexible and printed sensors and electronics draw much more attention to this topic [13], [14], and the priority of using printed electronics for sustainability is highlighted by technical committee 119 of the International Electrotechnical Commission who are developing standards for printed electronics [15].

Improved approaches for flexible biodegradable electronics are thus an active area of investigation. For example, Hosseini et al. [16] comprehensively reviewed biodegradable materials for health monitoring that can dissolve and degrade in vivo. In the review article by Sanchez-Duenas et al. [17], a wide range of biodegradable conductive inks and substrates have been investigated, with Mg and Zn considered popular for metal-based inks used for bioresorbable (materials to be fully degraded in vivo) electronics [18]. Similarly, Cook et al. [19] present Zn tracks on a transient substrate. Eco-friendly carbon-based inks and organic polymer-based inks have gained wide research interest [20]. Natural or synthetic polymers that can be mechanically, thermally, chemically, or biologically degraded are promising candidates for sustainable flexible substrates [21]. Recent studies on biodegradable wearable electronics studied the use of sustainable materials on wearable electronics such as sensors [22], field-effect-transistors [23], and Internet of Things (IoT) systems [24]. Kwon et al. [25] applied biocompatible, solderable graphene to multilayered electronic circuits. Highly conductive graphene has also been formed and patterned on natural woods and leaves [26]. Degradable and recyclable carbon-based electronics have been demonstrated in [27], and [28] overviews the life-cycle assessment of printed electronics.

The constituent parts of a complete flexible electronic system are shown in Fig. 1, with at least ten different aspects that enable flexible electronic devices. Hybrid integration of printed electronics combines conventional microchips and passive components with (generally) plastic substrates such as polyethylene naphthalate (PEN) and PET via printed silver tracks to form interconnections between components [29]. A sustainability challenge of integrated printed electronics is related to the recyclability of silver at the end-of-life [30]. Accordingly, an important open research problem is replacing metal interconnects with nonheavy metal materials. For instance, carbon-based interconnects have been presented in several works [31], but generally for the low-frequency aspects of a system, such as analog parts. At higher frequencies, parasitic capacitances and inductances can have a much larger impact than at lower frequencies. In a practical complete sensor system, digital microchips typically communicate with one another over a MHz range digital bus, such as serial-peripheral interface (SPI) or inter-integrated circuit (I2C). Indeed, it is increasingly common for sensor chips such as accelerometers to output digital data directly. It is therefore essential to investigate the effectiveness of using carbon-based interconnects in high-frequency digital communication channels. In this work, we aim to achieve this goal by investigating flexible biodegradable substrates paired with screen-printed carbon-based conductive tracks.

This article investigates the effectiveness of using screen-printed flexible carbon-based interconnects in digital communication systems. The output of this work supports the integration of biodegradable substrates in digital electronics systems. We investigate six commercially available sustainable substrates with conductive carbon-ink tracks while being used to carry SPI bus signals facilitating the communication between a micro-controller unit (MCU) and an electrochemical front end (EFE) unit capable of performing different types of voltammetric, amperometric, and impedometric measurements which may be used in future wearable sweat sensing systems such as [32] and [33]. We evaluate the printability, thermal properties, ink electrical performance, and mechanical durability of the screen-printed connections and provide guidelines for fabricating flexible interconnects suitable for digital electronic systems.

The remainder of this article is structured as follows. Section II describes the materials, manufacturing methods, and testing setup. Section III presents the experimental evaluation processes and results covering our implemented interconnect's printability, thermal stability, electrical performance, and mechanical performance. The findings are discussed in Section IV and the conclusions are then drawn in Section V.

II. METHODOLOGY

A. Substrate Selection and Track Printing

To investigate the effectiveness of using carbon-based tracks on different biodegradable substrates, we selected a set of commercial substrates. These include common packaging materials such as Nativia,[1] Natureflex,[2] and Greaseproof paper, healthcare sector materials like ClearFilm, and substrates that have been used in flexible electronics systems, such as cotton fabric [34] and Ecoflex [35] where biodegradability data has been reported previously in the literature. The selected substrates are listed below.

1) Nativia (Transpack, Southampton, U.K.).
2) Natureflex (Transpack, Southampton, U.K.).
3) Greaseproof paper, (Nisbets, Bristol, U.K.).
4) ClearFilm (Richardson Healthcare, Elstree, U.K.).
5) Cotton fabric (Premier Textiles, Stockport, U.K.).
6) Ecoflex (Smooth-On, PA, USA).

In addition to the substrates listed above, we used a 25-m-thick PEN substrate (Pütz Folien, Germany) for comparison. PEN is generally defined as being recyclable [36], meaning that the plastic waste material can be processed and reused for the original or other purposes. However, PEN is not considered biodegradable as it does not undergo significant

---

[1] Registered trademark.
[2] Trademarked.

TABLE I

INFORMATION OF THE SUBSTRATES USED IN THIS STUDY, AND WHETHER THEY ARE RECYCLABLE, BIODEGRADABLE, OR COMPOSTABLE

Substrate	Materials	Thickness (μm)	Stretchable	Breathable	Recyclable	Biodegradable	Compostable
PEN	Polyethylene naphthalate	25	No	No	Yes	No	No
Nativia®	Polyactic acid	35	No	No	Yes	Yes	Yes
Natureflex™	Wood pulp	30	No	No	No	Yes	Yes
Greaseproof paper	Paper pulp	80	No	No	No	Yes	Yes
ClearFilm (wound care)	Thermal plastic polyurethane	50	Yes	Yes	Yes	No	No
Cotton fabric	Cellulose fibres	250	Yes	Yes	Yes	Yes	Yes
Ecoflex	Silicone rubber	1500	Yes	No	Yes	Yes	Yes

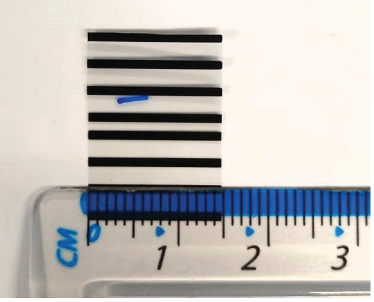

Fig. 2. Sample of the printed flexible digital interconnect with carbon-based tracks. Track length 1.5 cm, width 1 mm, and center-to-center pitch 3 mm (except the central tracks with a pitch of 2 mm).

chemical changes due to biological activity such as enzymatic action. It is also not compostable, meaning that it is not broken down by microorganisms under controlled conditions to produce stabilized organic residues, carbon dioxide, and water, either in the presence or absence of oxygen. The properties of our selected substrates are shown in Table I and reflect different options for being recyclable, biodegradable, and/or compostable. Plastic films with thicknesses ranging from 25 to 50 μm were selected. However, the thickness of the greaseproof paper and cotton fabric used in the study varies according to the manufacturer.

Most substrates were used as provided by the manufacturer with no further modifications. Backing papers, when present, were kept during the printing process and then removed for testing of the final interconnectors. The Ecoflex silicones were created as follows. A 1A:1B ratio was mixed, degassed, and cured overnight at room temperature (∼23 °C) to create a uniform substrate film.

Conductive tracks were created using screen-printed carbon ink, as shown in Fig. 2. The samples were derived from design patterns of our previous work in [32] and [33] and were used as straight interconnects to link different units via an SPI interface, simulating the interconnect between different components on a complete flexible PCB. The carbon ink used in this study, Dupont 7102, was selected for its nonheavy metal-based carbon material. However, we note that commercial inks commonly include nonbiodegradable additives to improve printability, flow, shelf life, and overall performance. As we focus on substrate characterization, we do not include a full characterization of this commercial ink. Rather, the choice of a commercial ink was made to simplify the experimental process and ensure reliable results. Before a complete system is used, all aspects in Fig. 1, not only the substrate, should be investigated for their sustainability profile, but this is beyond the scope of the current work.

Interconnects were fabricated using a TWS SR2700 semi-automatic screen printer provided by Blundell Production Equipment, Coventry, U.K. Tracks were cured at 120 °C for 10 min in a laboratory fan oven, adhering to Dupont 7102 carbon ink curing guidelines. This curing process is essential to enhance the electrical performance of the printed tracks. The screens used were sourced from MCI Precision Screens Ltd., U.K., with a mesh specification of PET 1500 120-34. The mesh parameters were chosen to balance printing resolution and layer thickness. While lower mesh counts could allow for single, thicker layers, they may compromise the resolution required for precise interconnect patterns.

In total, carbon ink was printed onto seven flexible substrates (those in Table I). For multilayer prints, the process was repeated, with additional layers carefully deposited over the previously cured ones. Fiducial markers were incorporated into the masks to ensure precise alignment during multilayer printing. This multilayer approach ensured uniform deposition while maintaining the fine details necessary for reliable high-frequency SPI communication.

B. Substrate Characterization

To characterize the fabricated interconnects, we utilized a scanning electron microscope (SEM) (TESCAN VEGA3, TESCAN, Czech Republic) to examine the morphology of carbon ink deposition on different substrates. Differential scanning calorimetry (DSC 250, TA Instruments, USA) was used to identify phase transition temperatures within the substrate materials (ranging from 20 °C to 300 °C at a scan rate of 10 °C/min). This process aimed to assess the thermal stability of the substrates during the ink-curing process. The thickness of the ink tracks was measured using a DektakXT[3] stylus profilometer (Bruker, Germany). DC resistance measurements were performed using a standard laboratory multimeter. For these measurements, five samples were used. The electrical impedance of the fabricated flexible interconnects was evaluated using two different devices to cover different frequency ranges: 1) Zurich Instruments MFIA, sweeping frequencies from 1 to 50 kHz; and 2) 1260A impedance analyzer (Solartron Analytical, U.K.) sweeping

---

[3] Trademark.

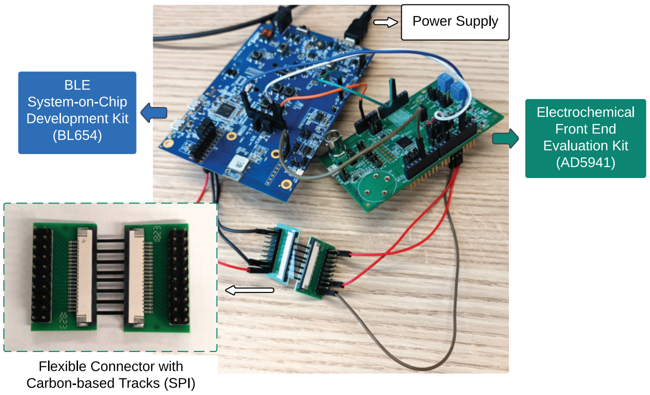

Fig. 3. Evaluation setup used to send SPI data between digital boards over the flexible interconnect on the biodegradable substrates.

frequencies from 100 kHz to 20 MHz. The presented results are the average from three samples.

C. Evaluation in Digital Communication Systems

An evaluation setup was implemented to test the effectiveness of using the fabricated flexible interconnects in digital communication systems (see Fig. 3). The setup was based on screen-printed carbon-based parallel tracks of 1 mm width and 15 mm length reflecting typical track sizes used on miniature printed electronics boards. These were on a horizontal pitch (center-to-center) of 3 mm, allowing them to be connected directly to a standard flexible flat cable (FFC) connector, connecting the interconnect under test to the test equipment. Two FFC connectors mounted on FR-4-based PCBs connected to the edges of the test printed unit and were wired to the other parts of the system.

The carbon-based tracks were used as an interconnect between two digital modules through an SPI bus. The SPI connection is based on the following signal terminals: main output secondary input (MOSI), main input secondary output (MISO), and clock (CLK). These were used to establish the SPI connection between two development boards: the DVK-BL654 (Laird connectivity, Akron, OH, USA) which supports applications for Bluetooth low energy (BLE), and the EVAL-AD5941 (Analog Devices, Wilmington, MA, USA) which facilitates the configuration and testing of an EFE AD5941 module.

The electronics evaluation setup aimed to emulate the use of flexible interconnects in high-frequency digital embedded systems. For this purpose, we programmed the AD5941 module to conduct differential voltage measurements between two internal voltage references: one set at 1.82 V and the other at 1.11 V. The BL654 module was configured to issue configuration and functional commands to the AD5941 module and to retrieve the results of the differential voltage measurement through the SPI port operating at MHz range frequencies. Ideally, 0.71 V should be measured as the difference between the two internal AD5941 references. These measurements were continuously transmitted to a PC via a USB port and displayed using a serial terminal application. Multiple samples of flexible interconnects were tested to establish a successful connection between the two modules. The success evaluation process was based on checking if the correct differential voltage measurement (0.71 V) was correctly displayed on the PC monitor.

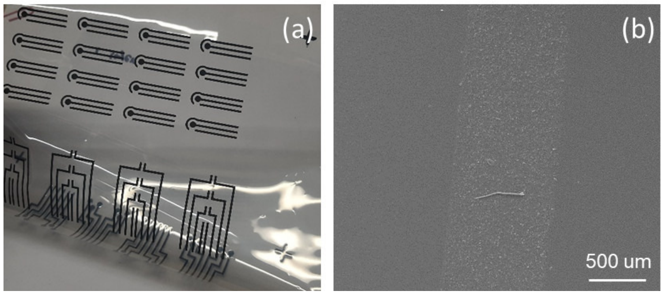

Fig. 4. (a) Optical and (b) SEM images of screen-printed carbon ink on a PEN substrate after curing at 120 °C.

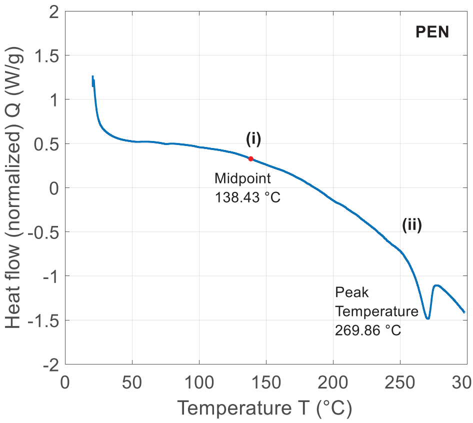

Fig. 5. Heat flow curve of PEN substrate under DSC from 20 °C to 300 °C, with (i) glass transition temperature and (ii) melting temperature noted.

III. EXPERIMENTAL EVALUATION

A. Printability and Thermal Stability of Substrates

To achieve high-quality, printed, electrically conductive tracks, the substrates must meet the ink curing specifications and offer suitable conditions for ink fixation, without deforming, decoloring, or losing their flexibility. As a baseline, the PEN substrate demonstrated in our previous work [32] was evaluated through optical and SEM imaging as shown in Fig. 4. Optical and SEM images in Fig. 4 revealed excellent ink fixation and smooth carbon ink tracks. The DSC curve shown in Fig. 5 revealed that the substrate has a glass transition temperature (Tg) of approximately 130 °C and a melting temperature (Tm) of around 260 °C. The two temperatures exceed the applied ink curing temperature of 120 °C. This ensures that the substrate has the required thermal stability during the ink-curing process.

As shown in Fig. 6, the same characterization process was applied to the six sustainable substrates. Smooth ink fixation was observed on Nativia [Fig. 6(a) and (b)], Natureflex [Fig. 6(c) and (d)], and greaseproof paper [Fig. 6(e) and (f)].

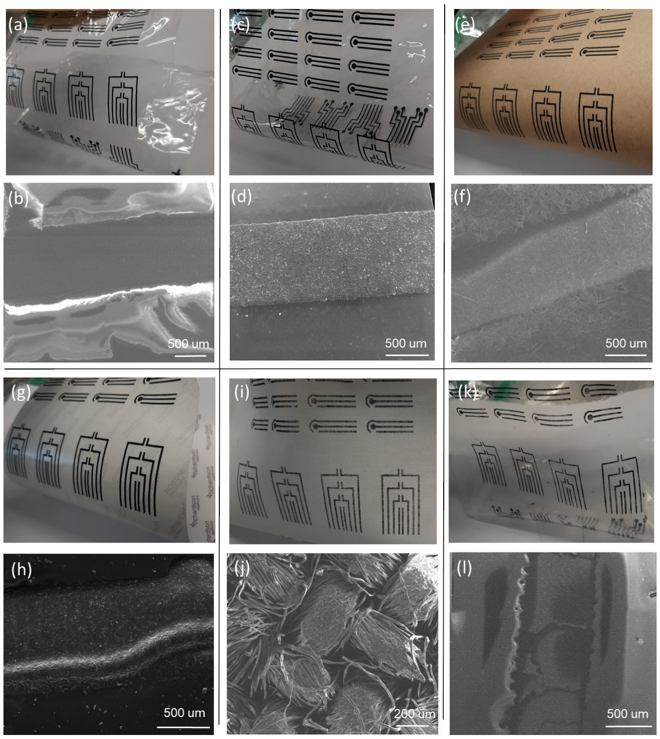

Fig. 6. Optical and SEM images of carbon ink cured on: (a) and (b) Nativia, (c) and (d) Natureflex, (e) and (f) greaseproof paper, (g) and (h) ClearFilm, (i) and (j) cotton fabric, and (k) and (l) Ecoflex.

However, wrinkles appeared on ClearFilm polyurethane (PU) film [Fig. 6(g) and (h)] when the PU layer deforms on top of the backing paper layer. In addition, we observed broken ink tracks on cotton fabrics [Fig. 6(i) and (j)] due to the uneven surface properties of the fabric, where the intersections between the yarns created difficulties for ink deposition. The Ecoflex substrate presented strong tackiness on the polyester screen mesh during the screen printing process, and broken carbon ink traces were found under SEM images [Fig. 6(k) and (l)], caused by the substantial flexibility of the substrate due to its low hardness.

Even with appropriate ink fixation, the thermal stability of the substrates remained a challenge for the sustainable substrates. This is because these substrates are designed to degrade easily under various conditions. As shown in Fig. 7(a), the low glass transition temperature (Tg) and melting temperature (Tm) of the Nativia substrate causes it to shrink and deform after curing at 120 °C for 10 min. In the heat flow curve of Natureflex [Fig. 7(b)], Tg is at 110 °C with a decompose temperature (Td) at around 235 °C. These temperatures reflect the observed substrate deformation under heat treatment for ink curing. Greaseproof paper demonstrated good thermal stability [Fig. 7(c)], where the phase transition occurred at 150 °C. The melting point of ClearFilm thermoplastic polyurethane (TPU) is lower than the typical value reported in the literature [36]. This discrepancy could be due to the thermal instability of the adhesives on the TPU backing at 120 °C. This also led to wrinkles of TPU on the backing paper as shown in Fig. 6(j). The cotton fabric and Ecoflex substrates decompose at temperatures higher than the ink-curing temperature. This confirms their thermal stability under the required ink-curing conditions. Although a water content loss is observed in the cotton fabric at approximately 130 °C, this has an insignificant impact on its overall thermal stability.

Characterizing the ink's printability and thermal stability on sustainable substrates demonstrated that carbon ink was successfully and uniformly deposited on Nativia, Natureflex, greaseproof paper, and ClearFilm PU films. In contrast, significant challenges were encountered with ink adhesion and fixation on cotton fabrics and Ecoflex substrates. These limitations rendered the latter materials unsuitable for subsequent mechanical testing.

B. *Electrical Impedance and Mechanical Durability*

The real part (resistance) of the impedance of fabricated tracks on the four substrates, where carbon ink was appropriately deposited, is shown in Fig. 8(b)–(d), in comparison to the carbon printed on a PEN substrate [Fig. 8(a)]. Stable and consistent resistance values were achieved across frequencies ranging from 1 to 50 kHz. The same range of resistance values (1.5 kΩ–2.5 kΩ) was observed for a single layer of screen-printed carbon ink on the different substrates.

In addition to the reference resistance values measured with the substrates in a flat posture, Fig. 8 illustrates the resistances corresponding to static mechanical deformations applied to the substrates, including bending (yellow) and stretching (red). These deformations were performed by curving the substrates over a beam with a radius of 1.7 mm, spanning an arc angle of ±180 angle. Bending is the configuration in which the printed carbon track faces inward toward the beam's center, while stretching occurs when the printed track faces outward.

The results revealed that bending the substrates reduced the electrical resistance of the tracks, whereas stretching increased it. This behavior can be attributed to the substrate's mechanical movement: bending mechanically compresses the carbon ink flakes, enhancing conductivity, while stretching disrupts the conductive paths, reducing ink connectivity. It is important to note that bending had a minimal impact on the measured resistance values in absolute terms. However, substantial differences were observed among the four types of substrates tested.

For example, during stretching, the most flexible substrate (ClearFilm PU film) exhibited the largest increase in resistance. This finding underscores the importance of investigating the mechanical durability of flexible substrates, particularly for use as tracks in electronic systems such as wearable bio-electronics, where physical flexibility is an important requirement of the overall system.

C. *SPI-Based Evaluation and Track Resistance Control*

To assess the effectiveness of using carbon-based flexible interconnects for SPI digital communications applications, we used the experimental setup shown in Fig. 3. While performing differential voltage measurements, we varied the SPI clock frequency to transmit digital data via the flexible tracks to the BL654 development board. Initial tests revealed that the

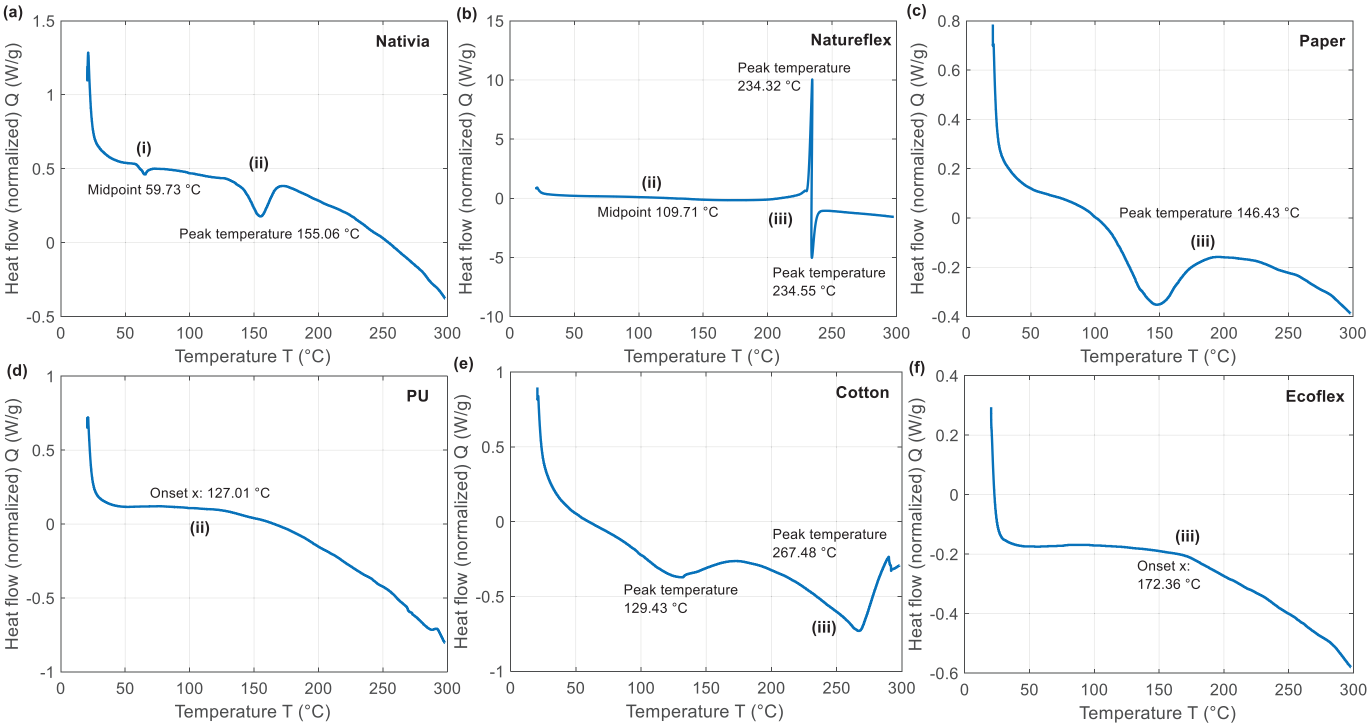

Fig. 7. Heat flow curves of (a) Nativia, (b) Natureflex, (c) greaseproof paper, (d) ClearFilm, (e) cotton fabric, and (f) Ecoflex, substrates under DSC from 20 °C to 300 °C, with their (i) glass transition temperature, (ii) melting temperature, and (iii) decomposition temperature marked. Note that degradation points are marked instead of melting points in Natureflex, greaseproof paper, cotton fabric, and Ecoflex substrates because the chemical decomposition process occurs before their phase changes.

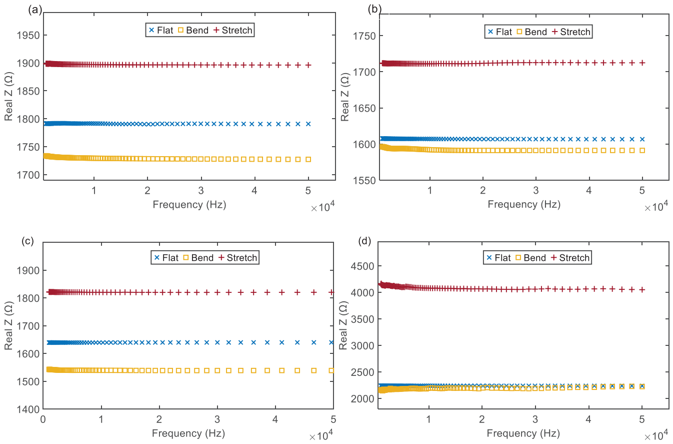

Fig. 8. Measured real impedance of one-layer of carbon ink printed on (a) PEN, (b) Natureflex (c) greaseproof paper, and (d) ClearFilm PU substrates. The substrates are placed in flat (blue), bent (yellow), and stretched (red) positions.

TABLE II

ELECTRICAL RESISTANCE NEEDED TO ENABLE ROBUST SPI COMMUNICATIONS AT DIFFERENT CLOCK FREQUENCIES

Freq. (MHz) / Res. (kΩ)	1	2	4	6	8	16
4.7	✗	✗	✗	✗	✗	✗
3.3	✓	✗	✗	✗	✗	✗
2.2	✓	✓	✗	✗	✗	✗
1.0	✓	✓	✓	✓	✗	✗
0.68	✓	✓	✓	✓	✓	✗
0.47	✓	✓	✓	✓	✓	✓

TABLE III

TRACK THICKNESS AND RESISTANCE VERSUS NUMBER OF PRINTED LAYERS ON A PEN SUBSTRATE. (MEAN ± STANDARD DEVIATION)

Printed layers	1 layer	2 layers	3 layers	4 layers	5 layers
Ink thickness (μm)	2.5 ± 0.2	5.9 ± 0.6	7.1 ± 0.3	8.6 ± 0.9	10.8 ± 0.8
Resistance (kΩ)	1.61 ± 0.10	0.86 ± 0.08	0.64 ± 0.04	0.51 ± 0.03	0.35 ± 0.03

electrical impedance of the conductive tracks disrupted SPI communication at higher frequencies. Therefore, we decided to replace the flexible test tracks with a series of resistors of known resistance to quantitatively analyze the impact of track electrical resistance on SPI communication across different frequencies. The results of these resistor-based experiments are shown in Table II. These can then be compared to the achieved printed resistances when using different numbers of printing layers, given in Table III.

We found that a resistance value of 3.3 kΩ allowed our SPI-connected units to communicate accurately at 1 MHz. However, for the devices to function properly at 16 MHz, the resistance must be reduced to below 680 Ω. This result leads to the conclusion that the SPI clock frequency and the

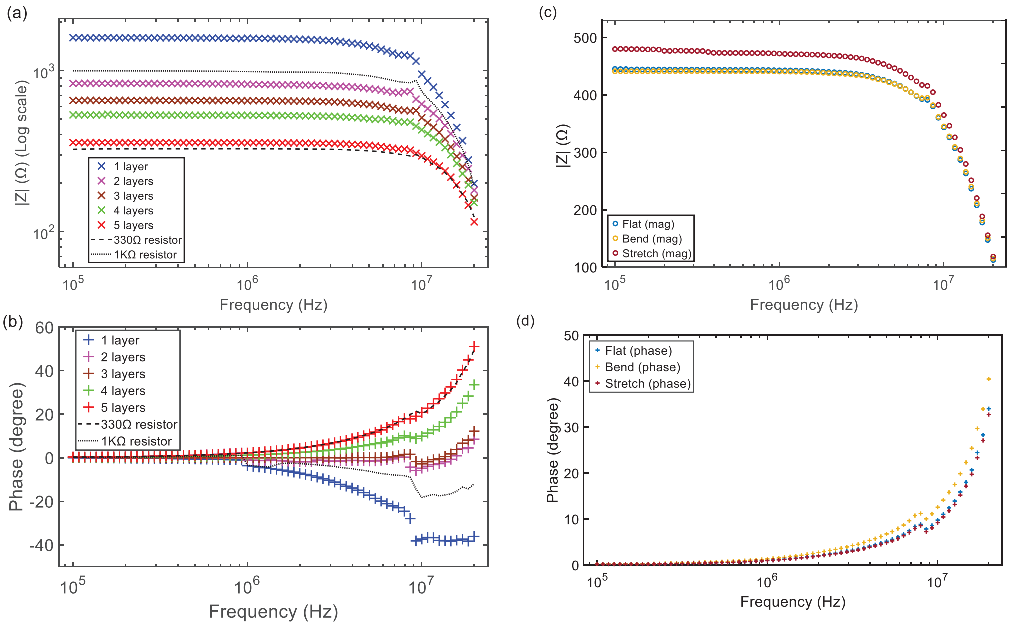

Fig. 9. Bode plot of impedance (a) magnitude and (b) phase of printed carbon ink on PEN substrates (from one to five layers of printing). The control resistors are off-the-shelf discrete component resistors with values 330 Ω and 1 kΩ for comparison purposes. Measured impedance (c) magnitude and (d) phase of Natureflex substrate screen-printed with three layers of carbon ink. The substrate is placed in flat (blue), bent (yellow), and stretched (red) positions.

resistance of the interconnection tracks are inversely related. Print quality thus trades off with the digital system design, and potentially other factors such as power consumption as communication at 1 MHz will take longer than communication at 16 MHz.

D. Resistance Control Through Multilayer Screen-Printing

The initial single-layer screen-printed carbon-based interconnectors were implemented using different flexible substrates and showed resistances ranging between 1.5 kΩ and 2.5 kΩ [see Fig. 8 (Flat)]. From Table II, this resistance range is unsuitable for implementing digital data transmission through SPI communication at high clock frequencies. Based on this, we decreased the resistance of the printed carbon tracks by using multilayer screen printing of the carbon ink. First, we evaluated this method by applying it to PEN substrates to find the optimal number of layers required for decreasing the electrical resistance. The results of these experiments (multilayer screen-printing of carbon ink on PEN substrates) are shown in Table III. Well-controlled ink thickness was achieved throughout the printing process. Thickness measurements were taken from five individual printed tracks, resulting in less than 1 μm standard deviations in all cases. The resistance values of these tracks (1.5 cm in length and 1 mm in width) had the largest standard deviation of 100 Ω in the one layer case. These results demonstrate how the thickness of cured ink tracks increases with each additional layer of printing, with a corresponding decrease in resistance.

As shown in Table III, the electrical resistance of the carbon tracks (measured using a multimeter) decreased as more layers were printed. Using three layers, the resistance dropped to < 680 Ω. Based on our previous experiments, this resistance range is considered suitable for SPI communication. However, as shown in Fig. 9 (Bode plots), when the frequency increased to more than 1 MHz, reactance effects begin to dominate, that is, frequency-dependent impedances due to effects of capacitance and inductance. At a frequency < 1 MHz, the printed tracks act as resistors in the Bode plot, maintaining constant magnitude and phase values. As frequency increases, we notice shifts in both magnitude and phase values. This indicates the effect of parasitic capacitance and inductance present in the printed tracks. (Control resistors, off-the-shelf discrete component resistors of 330 Ω and 1 kΩ, showed a similar impedance behavior across the frequency spectrum.)

The effect of parasitic capacitance was consistently observed across all samples of printed carbon-based interconnects at frequencies exceeding 10 MHz. As the frequency increased, higher conductivity interconnects (achieved through multilayer printing) showed larger shifts in magnitude and a positive phase shift. This indicates inductance in the circuit. Specific design guidelines should be followed to mitigate these effects, such as increasing the clearance between connection tracks and/or incorporating ground planes. In high-frequency communication devices like SPI buses, it is essential to maintain impedance matching across the Clock, MOSI, and MISO lines to minimize data interpretation errors and interruptions. These results highlight the importance of printing uniform carbon-based connection tracks to maintain precise impedance control in digital interconnects.

After implementing new carbon-based flexible interconnects using the proposed multilayer screen-printing method, we found experimentally that all interconnects made with three or more printed layers successfully provided stable SPI data communication at a clock frequency of 16 MHz. The carbon tracks on these interconnects have a length of 1.5 cm, width of 1 mm, and a horizontal center-to-center pitch of 3 mm. This result indicates that our proposed approach for interconnect design, maintaining suitable clearance distances (twice the width size), controlled ink thicknesses, and multilayer printing has effectively mitigated the effects of parasitics at SPI frequencies.

Among the six sustainable substrates considered in this study, Nativia, cotton fabric, and Ecoflex were excluded from the multilayer printing process due to their thermal instability or associated challenges during the printing process, as detailed in Section III-A. The opacity of greaseproof paper and ClearFilm PU substrates introduced difficulties in achieving precise alignment during the multilayer screen printing process, as their lack of transparency hindered visual guidance for layer alignment. In contrast, the transparency of the Natureflex film facilitated accurate alignment during multilayer printing, resulting in high-quality deposition of carbon ink with improved consistency and uniformity. This made Natureflex the most suitable substrate for demonstrating impedance measurements in the three-layer configuration. Fig. 9 shows the impedance measurements obtained from three-layer screen-printed carbon ink on Natureflex film. The measurements are similar to those obtained from measuring three-layer screen-printed carbon ink on PEN substrates. Mechanical bending and stretching had only a minor impact on SPI communication.

IV. DISCUSSION

This work has explored a range of sustainable substrates, including recyclable, biodegradable, and compostable options, to assess their suitability for printed flexible electronics. Interestingly, no direct correlation was observed between the disposal properties of these substrates and their printability or thermal stability. However, the successful demonstration of printable carbon interconnects on Natureflex film significantly expands the options of sustainable materials available for flexible electronic systems.

Natureflex was selected for the multilayer printing process due to its inherent transparency, facilitating precise alignment during the screen-printing process. This transparency enabled consistent layer stacking, ensuring high-quality deposition and uniformity of the carbon ink across the three layers. Such enhanced alignment reduced the occurrence of misalignment-induced defects, which were more prevalent with opaque substrates like greaseproof paper and ClearFilm PU. Moreover, Natureflex exhibited superior mechanical and thermal stability compared to the other tested substrates, minimizing deformation or ink delamination during the multilayer printing process. These attributes ensured that the three-layer printing process using Natureflex met the conductance requirements for reliable SPI communication.

It is important to note that the SPI tests conducted were limited to binary results, focusing on whether messages were correctly received. Consequently, no detailed investigation of the behavior of individual SPI lines at higher frequencies was performed, as this was outside the scope of the work. However, the high impedance in the system introduces parasitic effects, particularly at higher frequencies, which can degrade the integrity of SPI communication. A detailed investigation into these effects will be a valuable addition to future work, enabling the optimization of SPI communication performance in such systems. While Natureflex proved to be an excellent choice for multilayer printing in this study, the limitations of opaque substrates highlight an area for further development as they complicated the multilayer printing approach. While alternative methods, such as using a screen with increased layer thickness, can reduce resistance, multilayer printing was selected for its superior control over electrical properties and improved printing resolution. Future work to enable the broader use of sustainable opaque substrates like greaseproof paper and ClearFilm PU could expand the range of viable materials for flexible electronics, offering a balance between sustainability and performance.

In this study, we did not preheat the substrates before the printing was performed. This decision was made to maintain consistency across all substrate evaluations and to reflect typical conditions in real-world manufacturing processes. While preheating can help release inherent stresses and improve substrate stability under heat, our focus was on assessing the baseline performance of the substrates without additional processing steps. Future investigations could explore the potential benefits of preheating on substrate performance and its compatibility with carbon ink printing. Moreover, altering the ink composition to lower the curing temperature while maintaining high conductive performance could potentially broaden the substrate selection in this field of research; for reference, the ink used in this work was Dupont 7102 carbon ink, which requires curing at 120 °C for 10 min, as per manufacturer guidelines.

One potential challenge for printed carbon interconnects is their resistance to wear from repeated mechanical stress, particularly when they are used to make off-board connectors that are inserted and removed frequently. In this study, the FFC connector used in our test setup serves as an artifact of the test setup rather than a direct simulation of repeated insertions, as we did not aim to specifically evaluate the impact of mechanical stress in this experiment. However, it is important to note that, over time, the carbon layer might degrade with repeated friction from connectors, potentially affecting performance. Recent developments in portable impedance measurement systems have shown that continuous monitoring of impedance variations can provide critical insights into the electrical stability and performance of printed electronics under mechanical stress [37]. Such systems could play an important role in evaluating the durability and long-term reliability of printed connectors. It is important to explore this issue in more detail in future work. One method to address this limitation is to use protective coatings that could enhance the longevity of the connectors. Such encapsulation would likely be present in any end product/device, but is not part of our test structure in this article.

V. CONCLUSION

This study evaluated the printability and thermal stability of six commercial sustainable substrates. We identified Natureflex, greaseproof paper, and ClearFilm PU films as promising solutions to enhance sustainability in printed electronics. The electrical performance and mechanical durability evaluation of carbon ink screen-printed on these substrates provided insights into the suitability of different sustainable substrates. The effectiveness of using screen-printed carbon-based interconnects in digital communication systems was evaluated using a custom experimental setup employing a high-frequency SPI channel. Further, we introduced a multilayer screen printing technique to reduce the impedance of the carbon tracks, demonstrating that three layers of carbon ink printed on flexible substrates could be used for SPI communication. Overall, we highlight Natureflex as a flexible substrate for multilayer screen-printed carbon interconnects in high-frequency digital communication devices, as it has advantages such as transparency to enable accurate multilayer printing. Other evaluated substrates also hold potential as sustainable substrates, if specific challenges are addressed.

ACKNOWLEDGMENT

The data that support the findings of this study are available from the corresponding author, Alexander J. Casson, upon reasonable request.

REFERENCES

[1] P. Wang et al., "The evolution of flexible electronics: From nature, beyond nature, and to nature," *Adv. Sci.*, vol. 7, no. 20, Oct. 2020, Art. no. 2001116.

[2] Y. Wu, Y. Ma, H. Zheng, and S. Ramakrishna, "Piezoelectric materials for flexible and wearable electronics: A review," *Mater. Des.*, vol. 211, Dec. 2021, Art. no. 110164.

[3] A. Kothuru, C. H. Rao, S. B. Puneeth, M. Salve, K. Amreen, and S. Goel, "Laser-induced flexible electronics (LIFE) for resistive, capacitive and electrochemical sensing applications," *IEEE Sensors*, vol. 20, no. 13, pp. 7392–7399, Jul. 2020.

[4] O. A. Moses et al., "2D materials inks toward smart flexible electronics," *Mater. Today*, vol. 50, pp. 116–148, Nov. 2021.

[5] V. Beedasy and P. J. Smith, "Printed electronics as prepared by inkjet printing," *Materials*, vol. 13, no. 3, p. 704, Feb. 2020.

[6] W. Li et al., "Biodegradable materials and green processing for green electronics," *Adv. Mater.*, vol. 32, no. 33, Aug. 2020, Art. no. 2001591.

[7] R. Nithya, C. Sivasankari, and A. Thirunavukkarasu, "Electronic waste generation, regulation and metal recovery: A review," *Environ. Chem. Lett.*, vol. 19, no. 2, pp. 1347–1368, Apr. 2021.

[8] M. De Santis and I. Cacciotti, "Wireless implantable and biodegradable sensors for postsurgery monitoring: Current status and future perspectives," *Nanotechnology*, vol. 31, no. 25, Apr. 2020, Art. no. 252001.

[9] Y. Zhu et al., "A breathable, passive-cooling, non-inflammatory, and biodegradable aerogel electronic skin for wearable physical-electrophysiological-chemical analysis," *Adv. Mater.*, vol. 35, no. 10, Mar. 2023, Art. no. 2209300.

[10] B. N. Altay, M. Bolduc, and S. G. Cloutier, "Sustainable advanced manufacturing of printed electronics: An environmental consideration," in *Green Energy and Environment*, E. H. Yap and A. H. P. Tan, Eds., Rijeka, Croatia: IntechOpen, 2020.

[11] A. Sudheshwar, N. Malinverno, R. Hischier, B. Nowack, and C. Som, "Identifying sustainable applications for printed electronics using the multi-perspective application selection approach," *J. Cleaner Prod.*, vol. 383, Jan. 2023, Art. no. 135532.

[12] Y. Bonnassieux et al., "The 2021 flexible and printed electronics roadmap," *Flexible Printed Electron.*, vol. 6, no. 2, Apr. 2021, Art. no. 023001.

[13] Y. Luo et al., "Technology roadmap for flexible sensors," *ACS Nano*, vol. 17, no. 6, pp. 5211–5295, 2023.

[14] V. Pecunia et al., "Roadmap on printable electronic materials for next-generation sensors," *Nano Futures*, vol. 8, no. 3, Mar. 2024, Art. no. 032001.

[15] IEC Technical Committee 119. (2022). *Strategic Business Plan*. [Online]. Available: https://assets.iec.ch/further_informations/8679/sbp_119_2022-10.pdf?0906T07

[16] E. S. Hosseini, S. Dervin, P. Ganguly, and R. Dahiya, "Biodegradable materials for sustainable health monitoring devices," *ACS Appl. Bio Mater.*, vol. 4, no. 1, pp. 163–194, 2020.

[17] L. Sanchez-Duenas et al., "A review on sustainable inks for printed electronics: Materials for conductive, dielectric and piezoelectric sustainable inks," *Materials*, vol. 16, no. 11, p. 3940, May 2023.

[18] Y. Zhang, G. Lee, S. Li, Z. Hu, K. Zhao, and J. A. Rogers, "Advances in bioresorbable materials and electronics," *Chem. Rev.*, vol. 123, no. 19, pp. 11722–11773, Oct. 2023.

[19] A. Cook et al., "Sputtered Zinc electrodes on Pullulan substrates for flexible biodegradable transient electronics," in *Proc. IEEE Int. Conf. Flexible Printable Sensors Syst. (FLEPS)*, Boston, MA, USA, Jul. 2023, pp. 1–4.

[20] N. I. I. Maizal Hairi et al., "Recent advance in using eco-friendly carbon-based conductive ink for printed strain sensor: A review," *Cleaner Mater.*, vol. 12, Jun. 2024, Art. no. 100248.

[21] D. Gao, J. Lv, and P. S. Lee, "Natural polymer in soft electronics: Opportunities, challenges, and future prospects," *Adv. Mater.*, vol. 34, no. 25, Jun. 2022, Art. no. 2105020.

[22] J. Min, Y. Jung, J. Ahn, J. G. Lee, J. Lee, and S. H. Ko, "Recent advances in biodegradable green electronic materials and sensor applications," *Adv. Mater.*, vol. 35, no. 52, Dec. 2023, Art. no. 2211273.

[23] C.-J. Lee, Y.-C. Chang, L.-W. Wang, and Y.-H. Wang, "Biodegradable materials for organic field-effect transistors on a paper substrate," *IEEE Electron Device Lett.*, vol. 40, no. 2, pp. 236–239, Feb. 2019.

[24] H. Rahmani et al., "Next-generation IoT devices: Sustainable eco-friendly manufacturing, energy harvesting, and wireless connectivity," *IEEE J. Microw.*, vol. 3, no. 1, pp. 237–255, Jan. 2023.

[25] Y.-T. Kwon et al., "All-printed nanomembrane wireless bioelectronics using a biocompatible solderable graphene for multimodal human-machine interfaces," *Nature Commun.*, vol. 11, no. 1, p. 3450, Jul. 2020.

[26] T.-S.-D. Le, S. Park, J. An, P. S. Lee, and Y.-J. Kim, "Ultrafast laser pulses enable one-step graphene patterning on woods and leaves for green electronics," *Adv. Funct. Mater.*, vol. 29, no. 33, Aug. 2019, Art. no. 1902771.

[27] J. Szałapak et al., "Carbon-based composites with biodegradable matrix for flexible paper electronics," *Polymers*, vol. 16, no. 5, p. 686, Mar. 2024.

[28] T. M. Prenzel, F. Gehring, F. Fuhs, and S. Albrecht, "Influence of design properties of printed electronics on their environmental profile," *Matériaux Techn.*, vol. 109, nos. 5–6, p. 506, 2021.

[29] Y. Khan, A. Thielens, S. Muin, J. Ting, C. Baumbauer, and A. C. Arias, "A new frontier of printed electronics: Flexible hybrid electronics," *Adv. Mater.*, vol. 32, no. 15, Apr. 2020, Art. no. 1905279.

[30] A. Sudheshwar, N. Malinverno, R. Hischier, B. Nowack, and C. Som, "The need for design-for-recycling of paper-based printed electronics—A prospective comparison with printed circuit boards," *Resour., Conservation Recycling*, vol. 189, Feb. 2023, Art. no. 106757.

[31] Y. Qin, X. Ouyang, Y. Lv, W. Liu, Q. Liu, and S. Wang, "A review of carbon-based conductive inks and their printing technologies for integrated circuits," *Coatings*, vol. 13, no. 10, p. 1769, Oct. 2023.

[32] Z. Wang, A. Alwattar, P. Quayle, J. C. Batchelor, and A. J. Casson, "Effects of sensor design on the performance of wearable sweat monitors," in *Proc. IEEE Sensors*, Oct. 2022, pp. 1–4.

[33] M. Saleh, Z. Wang, J. C. Batchelor, and A. J. Casson, "A portable electrochemical measurement platform for wearable-flexible sweat sensors," in *Proc. IEEE Sensors Appl. Symp. (SAS)*, Ottawa, ON, Canada, Jul. 2023, pp. 1–6.

[34] Z. Wang, T. Lowe, and B. Derby, "Fluid/fiber interactions and the conductivity of inkjet printed Ag on textile substrates," *ACS Appl. Mater. Interfaces*, vol. 12, no. 40, pp. 45516–45524, Oct. 2020.

[35] S. Kim et al., "An intrinsically stretchable multi-biochemical sensor for sweat analysis using photo-patternable ecoflex," *npj Flexible Electron.*, vol. 7, no. 1, p. 33, Jul. 2023.

[36] J. Wiklund et al., "A review on printed electronics: Fabrication methods, inks, substrates, applications and environmental impacts," *J. Manuf. Mater. Process.*, vol. 5, no. 3, p. 89, Aug. 2021.

[37] M. Saleh, S. Medina-Lombardero, M. L. Crichton, and A. J. Casson, "A portable electrical impedance measurement system for flexible electrodes," in *Proc. IEEE SENSORS*, Kobe, Japan, Oct. 2024, pp. 1–4.