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Biodegradable and Nanocomposite Materials as Printed Circuit Substrates: A Mini-Review

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ABSTRACT Biodegradables are a promising path for the future of electronics in a greener mindset. The review study focuses on their applications and past and current research results. The paper also investigates the application of nanomaterials as fillers to control or increase the physical (electrical, mechanical, thermal) properties of biodegradable biopolymers. These biodegradables and nanocomposites are already effectively used in prototypes and advanced application areas with demanding requirements, such as flexible and wearable electronics, implantable or biomedical applications, and traditional commercial electronics. The nano-enhanced biopolymer substrates (e.g., with improved gas and water barrier functionalities) sometimes also with integrated, nano-enabled functionalities (such as electromagnetic shielding or plasmonic activity) can be beneficial in many electronics packaging and nanopackaging applications as well.

INDEX TERMS Printed circuit substrate, biodegradable materials, nanocomposites, nanomaterials.

I. INTRODUCTION

Electronics industry is facing significant problems with increasing production and, consequently, the problem of hazardous electronic waste [1].

To eliminate the dangerous materials contained in electronic waste, industries must develop new ideas and approaches. The issue is additionally elevated by the global market-based attitude, which encourages consumers to discard outdated hardware. While the majority of electronic equipment and modules are essentially non-degradable, proper recycling is also a crucial concern. Recently, some rules were implemented to prevent the manufacture of devices with hazardous elements (an example: the "RoHS Directive", which was a key action in this matter more than a decade ago [2], [3]).

Printed electronics have a long-running history in the research and development of electronics. The first patent for printed wires was filled by Hanson [4], where the dielectric was paraffined paper. The large-scale production of printed circuit boards (PCBs) was based on Eisler's technology [5], and commercial PCB technology was introduced in 1948 [6]. Bio-based and modern biodegradable materials only surfaced recently in the field of printed electronics in advance researches. In this field, we must separate printed electronics on plastics and flexibles from conventional PCB technologies involving rigid, multilayered PCBs. These substrates can be a promising alternative in the circular-economy-based pathfinding processes of the electronics industry, which is facing problems from the ever-emerging e-waste management requirements. Traditional electronics substrates raise huge environmental concerns, as they can be considered the fastest-growing category of hazardous solid waste in the world [7]. The reported amount of E-waste was around 50 million tons in 2020; the estimation for 2050 presents 120 million tons per year [8]. A possibility to decrease plastics in electronic products is to use materials where advanced end-of-life recycling and recapture can be used - in this vision, biodegradables form a potential future application.

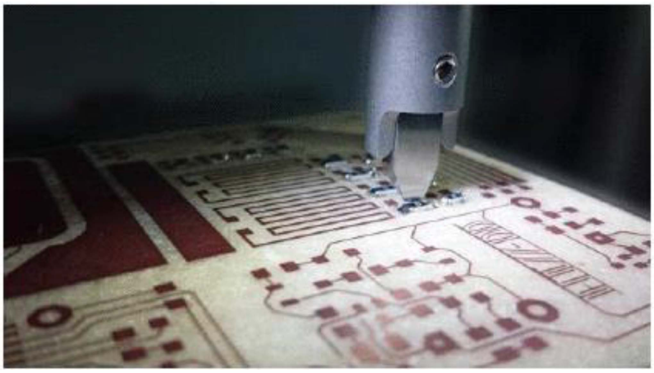

FIGURE 1. SMT component test on printed circuit board made of sustainable material [9].

Fig. 1 shows a recent example [9] of applying biodegradable substrate for traditional surface-mount technology (SMT) based PCB assembly.

Although biodegradable electronic circuit substrates have many favorable properties, modification of their mechanical, electrical, or thermal properties is often needed for advanced applications. By using nanomaterials as fillers, these physical properties can be controlled and improved to match the application-driven requirements. Moreover, new functionalities can also be enabled. The benefits of creating and using such nanocomposites in conjunction with the biodegradable substrates will also be discussed in Section III.

II. BIODEGRADABLES AS CIRCUIT SUBSTRATES

A. BIODEGRADABLES

Special types of polymers, known as bioplastics, become widely known with the development of amylomaize in the 1950s. Bioplastics can be biodegradable and compostable [10], [11] (in accordance with the DIN 12432 standard [12]), or they can be produced from renewable resources.

There are various subclasses of bioplastics [10], including:

- Not bio-based, but compostable or biodegradable
- Biobased, yet non-biodegradable;
- biodegradable or compostable at the same time.

Some publications separate biodegradability and compostability of plastics [13], but the overall specification is not set globally yet. Nägele et al. highlighted early [14] that not only the ecological aspects of renewable bio-resources are important in this field; they pointed out the possible increase in costs, which is due to the further depletion of fossil resources. These factors urge the spread of developing novel materials and technologies in the production of electronic substrates.

In terms of waste handling, printed circuit boards and printed circuit substrates are essential components of an electronic assembly. Still, the substituting of classical compositions (such as flame-retarted epoxies with glass fiber reinforcement) is not solved generally in the field. The usage of environmentally friendly materials in electronics is given in the literature as advanced research. Still, despite the beneficial qualities of such substrates, they are rarely used in consumer devices yet. The current findings are well below the technological readiness level of mass production.

It must be noted that the use of biodegradables as printed electronics substrates does not imply that these materials will break down during the course of their lifespan, but in specific composting processes or dissolution [15]. The key is that the procedure gets more straightforward and more environmentally beneficial as the item reaches the end of its useful life and becomes e-waste. However, bio-based and biodegradable materials are not always desirable for application. According to Adnan et al. [16], inorganic biodegradable substrates sometimes give better results than other types of biodegradables. It is also important to note that they referred to the substrates used in traditional electronic active device (e.g., IC packaging) processes, not in printed circuit electronics.

B. MATERIALS AND PROCESSES USED TO PRODUCE PRINTED ELECTRONICS ON BIODEGRADABLES

Abdulrhman et al. showed that polycaprolactone (PCL) could be used for circuitry integration, staying in the regime of flexible substrates [17]. Yedrissov et al. [18] very recently proposed a new method for producing polylactic acid (PLA)-based PCBs to replace environmentally hazardous polymer binders with a biodegradable solution. The production and the recycling process were presented, the proposed material with the recycling method aims to save and reuse the most valuable components of the PCB. PLA was also used by other researchers for composition of such devices. Géczy et al. showed PLA and cellulose-acetate (CA) boards could be used for surface mounting. The research group also produced several other publications, involving PLA and CA, as PCB substrate [19], [20], [21], [22], [23], [24], [25], [26], [27]. Recently a promising improvement was introduced by the same team, where flame-retarded PLA was reinforced with flame-retarded flax fibres, to form a biodegradable substrate [27]. Lincoln et al. [28] also tested flax-reinforced bioepoxies - the flammability, thermal resistance, mechanical performance, and electrical properties of prototype circuit boards. The bioepoxy-flax composites show promising results in terms of toxicity, biodegradability, energy consumption, GHG emissions, and economic costs compared to the petroleum-based, epoxy-fiberglass composites.

Henning et al. [29], [30], [31] compared ternary and binary PLA composites as printed wiring substrates. They were able to realize double sided wiring boards composed of PLA [29]. Veselý et al. [32] conducted measurements on 3D printed PLA test samples to evaluate mechanical and thermomechanical properties.

Guna et al. [33] applied biocomposites to develop biodegradable PCBs, using banana stems and wheat gluten to extract natural cellulose fibers. After the achievement of flame retardancy, adequate dielectric requirements, and performance stability, the proposed agricultural waste and coproducts are offering a feasible solution to produce biodegradable PCBs.

Bharath et al. built PCBs from rice husk fiber. They mixed the material with epoxy resin, which renders their composition only partially degradable [34].

Kumar and Gupta [35] conducted tensile test, 3-point bending test, thermal analyses, and dielectric analyses to examine the applicability of jute, hemp, and sisal in PCBs instead of glass fiber epoxy composite. The authors found the presented natural biocomposites suitable for PCB producing. However, the exact composition is not revealed in their paper.

Liu et al. [36] performed life cycle assessment study about the environmental impact of paper-based multilayer printed circuit boards. The paper examined the acidification potential, global warming potential, toxic potential, and ozone layer depletion potential. The environmental impact index is determined, which is found about two orders of magnitude lower than in the case of organic printed circuit boards. However, the examined solution is not suitable for high-density and high-performance integrated circuit applications.

Immonen et al. [37] investigated the possibility of cardboard and veneer as substrate materials for biodegradable printed circuit boards. The study examined print quality, electrical conductivity, fire performance, and biodegradation. The results verified the usefulness of wood-based substrates in environmental or medical analytics, but an adequate fire-retardant coating is necessary to avoid ignition due to electrical shortcuts.

Fang et al. [38] reviewed the application of versatile wood cellulose in electronics. The study collected information about cellulose substrates, versatile wood cellulose as functional components, and smart materials.

Arroyos et al. [39] presented a functional mouse prototype to demonstrate design principles for sustainable electronic devices. The mouse consists of a biodegradable printed circuit board (SoluBoard - as the commercialized title shows), compostable case, and minimal electronics. The authors reported that the design reduces the environmental carbon impact by 60.2% compared to a traditional mouse. However, the Soluboard assembly needs to be performed in reflow soldering (the prototype was made with hand soldering).

C. FLEXIBLE SUBSTRATES AND DEVICES

Next to the traditional PCB substrates, flexible and electronic device substrates are also investigated from this aspect. Daniele et al. [40] applied a citric acid-based elastomer, POMaC - Poly(octamethylene maleate (anhydride) citrate) - as PCB substrate and packaging. Single and multilayer layer POMaC-PCBs were produced and characterized. Swelling behaviour, degradation behaviour, tensile properties, and crosslinking conditions were tested.

Held et al. [41] investigated the mechanical properties of biodegradable and photo-cross-linkable elastomer poly (glycerol sebacic) acrylate (PGSA) as a component of soft and stretchable electronics. The rubber-elasticity, biodegradability, and conductivity properties make the examined platforms ideal for agriculture, healthcare, packaging, and disposable electronic consumer products.

Chandrasekaran et al. [42] used cellulose-laponite composite as substrate material for printed electronics. The authors examined the effects of laponite with various mass ratios. They found that the thermal and flame retardancy properties are increased by adding laponite into the cellulose, although it makes the substrate more brittle.

Jung et al. [43] showed that key electrical components could be presented on flexible cellulose nanofibril paper. Their results had comparable performance with the standard components.

Mattana et al. [44] employed polylactic acid (PLA) thin films as substrates for organic electronic devices. The authors reported that the examined substrates are applicable for disposable electronics after the mechanical and dielectric characterization. The study presents the fabrication of organic field-effect transistors (OFETs) and all-inkjet-printed organic electrochemical transistors (OECTs) on top of PLA.

D. ASSEMBLING

Regarding the assembly of components to biodegradable substrates, soldering is often applied. A low-temperature solder alloy based on bismuth usually is necessary because of the biopolymer low thermal resistance [18], [19], [29], [45]. As the study by Henning et al., 2019 [29] states, the solder joints exhibit a higher shear strength than those made by conductive epoxy-based adhesive. Nevertheless, the warping of the boards usually occurs due to high differences between the thermal expansion coefficients of the substrate, copper layer, and components. The mechanical properties of resulting joints are usually not as good as those made on FR4 substrates. [46] reported in their work that the most problematic aspect of the assembly process is heat transfer, which can lead to the delamination of the substrate. Several soldering techniques were investigated in the study by [19], including hot-iron, selective hot-gas, laser, and vapor phase soldering (VPS). They concluded that VPS is the most promising from the list, due to the uniform heating and applicability of a low-temperature heat-transfer medium. The shear tests revealed that the solder joints on PLA-based substrate prepared by VPS are almost comparable to those on FR4. Nevertheless, the quality of joints depends on the specific type of biopolymer [45]. Further, instead of standard bismuth-tin solder alloy (58Bi42Sn or 58Bi41Sn1Ag), Rose's alloy with the composition of 50Bi (25-28) Pb (22-25)Sn can be beneficially utilized thanks to its melting point of 94 °C [17].

Next to soldering, components mounting via electro-conductive adhesives (ECA) can be a feasible alternative to joining. A benefit of this technique is the possibility of adhesives curing at room temperature. Thus, thermal treatment impacting the assembly can be avoided [47]. In terms of weaker thermal resilience of biodegradable substrates, adhesives that can be also biocompatible and water-soluble need to be adopted. Huang et al. utilized polyethylene oxide (PEO) [48] filled with metal (tungsten and zinc) particles for both creating the conductive traces and mounting the electronic components. The paste showed acceptable mechanical properties for joining the components, but concerns regarding delamination of the conductive pads were reported. As a substrate, they utilized sodium carboxymethylcellulose sheets bonded together by PEO. Liu et al. used paper-based substrates [36] for PCB production. In their report, the conductive pattern was created using screen-printed polyurethane-based ECA, while the SMDs were assembled by application of the ECA based on polyacrylate. Compared to other organic PCBs, paper-based PCBs were found to have an excellent reliability performance from a thermal cycling point of view. On the contrary, the novel material is not resistant to humidity and mechanical stress and is applicable for transmitting signals of high frequencies. Substrates made of the biopolymers consisting of polylactic acid (PLA) and thermoplastic polyester elastomer (TPC) were prepared by Schramm et al. [45]. Mounting of the SMDs was performed by ECA consisting of epoxy-based binder and silver filler. The study includes the assessment of the implementation hot embossing process to arrange the conductive pattern and the evaluation of the humidity effect. Results demonstrated concerns with the delamination rather than with conductive joints. A different manner of PCB fabrication was utilized by Abdulrhman et al. [17] and Guna et al. [33], who first deposited silver on the biodegradable substrate, and then the silver was covered by copper. In the work of Abdulrhman, a substrate was prepared of polycaprolactone (PCL). Guna et al. established a composite composed of banana fibers and wheat gluten. In both cases, the components for testing the functionality of the circuit were mounted using ECA. The testing did not reveal any defects in the circuit, even after several days of service. A direct comparison of adhesive and soldering means of components mounting on the biodegradable substrate was conducted by Staat et al. [9]. Moreover, the paper comprises an investigation of various finishing processes concerning the pattern formation on the PLA substrate.

E. RELIABILITY AND QUALITY ISSUES

Bozó et al. [49] highlighted that bioplastics could be employed during the manufacturing of microelectronics components and devices, but compared to the conventional polymers, the results show inferior thermal and mechanical properties. Most papers cite this as a general problem with biodegradables in electronic packaging.

Silver, contained in the conductive adhesives, is prone to electrochemical migration. This reliability issue occurrence could also be affected by the type of substrate. The biodegradable substrates differ in surface structure from the traditional FR4 substrates and exhibit different wetting characteristics of liquids. This fact, combined with the lower accuracy of the conductive pattern shape, could lead to higher susceptibility to electrochemical migration [50]. Biopolymers can be used as packaging material thanks to their high crystallinity and hydrophobicity [51]. Cao and Ulrich [52] found that the mechanical flexibility of synthetic polymers can be adjusted in the case of stretchable biodegradable electronics.

TABLE 1. Material Properties of Commonly Used Biodegradable and Biocompatible Substrate Materials

Polymer matrix	Biodeg/ Biocomp	Conductivity [Scm^{-1}]	UTS [MPa]	T_g [°C]	Refs.
Cellulose / CA	yes/yes	10^{-13}–10^{-11}	10–130 (films); 110–1430*	200–250	[53-56]
Collagen / Gelatin	yes/yes	6.5×10^{-12}	50–100	30–60	[57-59]
PDMS	no/yes	2.5×10^{-16}	2.3	150	[60-61]
PGSA	yes/yes	10^{-9}–10^{-8}	0.1	28–32	[41, 61-63]
PLA	yes/yes	4.2×10^{-12}	49.3	54.1	[64-65]
PLGA	yes/yes	9.8×10^{-11}	8-15	45–65	[66-69]
PVA	no/yes	1.63×10^{-12}	4–10.5	31–49.4	[70-72]
Shellac	yes/yes	1.25×10^{-8}	12–33	41–49	[73-75]
Silk	yes/yes	1×10^{-15}	500–1500	130–243	[76-78]

*nanocellulose derived

Table 1 summarizes the material parameters for the most common substrates: biodegrability or compostability, conductivity [Scm^{-1}], UTS – ultimate tensile strength [MPa], and T_g – glass transition temperature [°C].

III. NANOCOMPOSITES

Biopolymer nanocomposites are materials composed of biopolymer matrices modified with nanoscale filler materials. While in bulk nanocomposites, these nanofillers are dispersed evenly in the polymer matrix, in the case of surface nanocomposites, the nanomaterials are usually layered on the surface of a polymer or segregated in a subsurface layer [88]. The role of nanomaterials is twofold, they can either improve the physical properties of the matrix material (nano-enhanced applications), or they can enable new properties and functionalities (nano-enabled applications).

For both purposes, the substrate materials discussed in the previous section (and listed in Table 1) can be considered ideal biopolymer matrices for nanocomposite fabrication. The addition of different nanomaterials (metallic nanoparticles and wires, metal-oxides, carbon allotropes, etc.) usually does not impair the biodegradable/ biocompatible properties of the substrates. In contrast, the mechanical, electrical, optical, thermal, etc. properties could be influenced to a great extent. Considering printed circuit substrates, we will concentrate on the most relevant properties regarding this application. For readers interested in a wider application area of nanocomposites in electronics, several in-depth reviews can be recommended [51], [89], [90]. Table 1 presents the material properties of common conductive nanocomposite materials based on biodegradable or biocompatible matrixes.

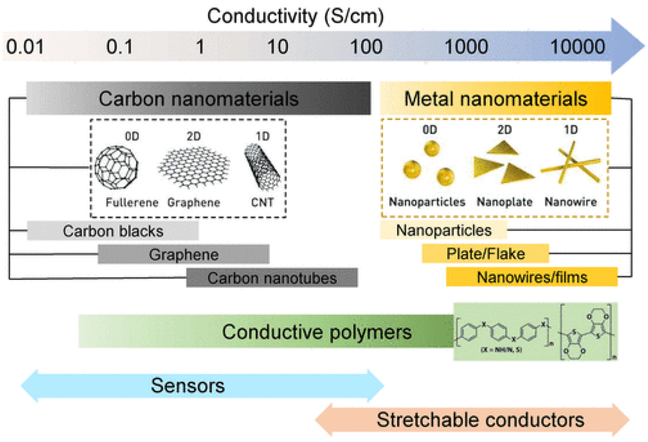

FIGURE 2. The conductivity of different nanomaterial fillers and nanocomposites with their potential application areas. Reprinted with permission from [92]. Copyright American Society of Chemistry 2019.

A. ELECTRICAL PROPERTIES

Although the primary purpose of using biodegradable substrates is E-waste management, many advanced applications require the modification of the electrical properties of the matrix materials, especially for flexible substrates, where matching the mechanical properties of the substrate and conducting layers is important (e.g., stretchable electronics, wearable or implantable biomedical applications, etc.) [91], [92]. As illustrated in Fig. 2, a variety of nanofillers, mostly carbon allotropes and metallic nanomaterials can be used to create conductors from the generally insulating biopolymer materials or increase the conductivity of the few conductive/semiconductive polymers (e.g., polyaniline (PANI), polypyrrole (PPy), Poly(34-ethylenedioxythiophene), (PEDOT)) as well [51].

Table 2 presents a few examples of conductive nanocomposite materials based on the most frequently used biodegradable and biocompatible biopolymer matrices. As a general rule of thumb, anisotropic metallic nanomaterials (such as nanowires or nanoflakes) provide the best conductivity at the lowest filler contents [87], which is important for maintaining the favorable mechanical properties of the substrate as well. Heterostructures, combining nanomaterials with different dimensions [86], or surface nanocomposites [83] are also effectively used to tackle this issue.

On the other hand, nanomaterials can also be used to improve the dielectric properties of substrate materials for electrical energy storage applications. For example, Lao et al. used 10wt% boron nitride nanosheets (BNNS) to create dielectric polymer films with an energy storage density of 4.1 $\mathrm{Jcm^{-3}}$ and a breakdown voltage of 370 MV $\mathrm{m^{-1}}$.

B. MECHANICAL PROPERTIES

A favorable property of many substrate materials listed in Table 1 is their flexibility, which can be advantageous both during processing/manufacture or during applications, such as in bendable, stretchable, or wearable electronics. Although the Young's modulus and tensile strength of a nanocomposite are primarily determined by the matrix, the nanofillers have an influence on it, and in most of the cases, care should be taken not to decrease the flexibility while, for example, creating a conducting nanocomposite [92].

TABLE 2. Material Properties of Common Conductive Nanocomposite Materials, Based on Biodegradable or Biocompatible Matrixes

Matrix	Biodeg / Bioc.	Dopant	Conduct. [$\mathrm{Scm^{-1}}$]	UTS MPa	Ref.
Cellulose / CA	yes/yes	MXene ($Ti_3C_2T_x$) + 20% CNFs	2.95×10^{2}	341	[79]
Collagen	yes/yes	10% Cu NPs	1×10^{-3}	N.A.	[57]
		2 wt% FWCNT	4.8×10^{-4}	16	[80]
		2 wt% BCNT	3×10^{-1}	19	[80]
Gelatin	yes/yes	graphene (rGO)	4.3×10^{2}	630	[81]
PEDOT	no/yes	SCNT–PG–PEDOT network	1.88×10^{2}	31.6	[82]
PANI	no/yes	0.84 mg $\mathrm{cm^{-2}}$ Ag NW (on surface)	1.03×10^{4}	N.A.	[83]
PCS	yes/yes	30% CSNWs	5×10^{-4}	11	[84]
PDMS	no/yes	28 wt% GNP	1×10^{-1}	N.A.	[85]
		3D PS microspheres, 2D RGO nanosheets, 1D AgNWs	1.23×10^{1}	N.A.	[86]
PLA	yes/yes	5wt% CNT	3.6×10^{-3}	68.7	[64]
		5wt% CNT, 0.1wt% graphene	1.32	69.3	[64]
PSBS	no/yes	ultralong gold-coated silver nanowires	4.2×10^{4}–7.3×10^{4}	2	[87]
PVA	no/yes	0.4 wt% GO	1.4×10^{-10}	N.A.	[71]
Silk	yes/yes	graphene	1.4×10^{-3}–1.1×10^{-2}	80-90	[77]

For abbreviations see the list at the end of the paper.

Balancing the mechanical and electrical properties is also important for stiff printed circuit substrates. Significant improvement in the yield strength, elongation at yield, Young's modulus, and toughness properties were demonstrated with the addition of carbon nanomaterial fillers, e.g., with 5 wt% CNT in a PLA matrix [64].

C. THERMAL PROPERTIES

For high-power, flexible electronic applications, substrates that can effectively dissipate heat while maintaining their mechanical stability are required. Here the aim of doping is to increase the thermal conductivity of the substrate while maintaining its mechanical flexibility. Carbon-based nanofillers are especially promising for this purpose since they combine good thermal conductivity (from $\sim$2000 Wm^{-1}K^{-1} for graphene to $\sim$100 Wm^{-1}K^{-1} for bulk graphite [91]) with low density. By using graphite nanoplatelets, Burzynski et al. managed to increase the thermal conductivity of PDMS 9× to 1.8 Wm^{-1}K^{-1} at only 11 vol% filler content [85].

D. OTHER PROPERTIES

Considering electronics packaging applications, important factors can be to improve the gas and water barrier capabilities of the substrates. For this purpose, GO nanosheets were successfully used to decrease the oxygen and water vapor permeability of the polymer matrix [93]. Also, some special electronic devices need packaging with materials having electromagnetic shielding and antistatic functions. Conductive nanofillers, such as MWCNTs were successfully used to create an environment-friendly, biodegradable, low-weight PLLA/MWCNT nanocomposite foam for electromagnetic shielding applications [94].

Besides controlling the conductivity, metallic nanoparticles are often used to endow the biopolymer matrix with plasmonic or catalytic properties, which can be beneficial considering integrated sensor functions and applications [92].

IV. CONCLUSION

In the paper, we summarized and reviewed the most recent literature regarding biodegradables in printed electronic substrates.

With a discussion on biodegradables we established the general overview of the term and the materials (also the related processes) used to produce and assemble printed electronics on biodegradables. We also focused on flexible substrates and devices; furthermore, the reliability and quality issues were also summarized.

It was also demonstrated through examples that with the help of nanomaterial fillers, the most important physical properties (electrical, mechanical, and thermal) of biodegradable matrices could be controlled and improved. These nanocomposites can enable novel functionalities and tackle the requirements of advanced application areas.

The future shows promising application possibilities for such substrates. Still, while the presented demonstrators are mainly laboratory prototypes, the time is near when the technology readiness level will elevate from the general state of advanced research and prototyping.

LIST OF ABBREVIATIONS

BCNT:	boron-doped carbon nanotube; BNNS: boron nitride nanosheet.
CNF:	cellulose nanofibril.
CNT:	carbon nanotube.
CSNW:	copper sulfide nanowires.
ECA:	electroconductive adhesives.
EM:	electromagnetic.
FWCNT:	few-walled carbon nanotube.
GHG:	greenhouse gases.
GNP:	graphite nanoplatelet.
GO:	graphene oxide.
MWCNT:	multi-walled carbon nanotube.
NP:	nanoparticle.
NW:	nanowire.
OECT:	organic electrochemical transistors.
OFET:	organic field-effect transistors.
PANI:	polyaniline.
PCB:	printed circuit board.
PCL:	polycaprolactone.
PCS:	poly(citrates-siloxane).
PDMS:	polydimethylsiloxane.
PEDOT:	poly(34-ethylenedioxythiophene).
PEO:	polyethylene oxide.
PGSA:	poly(glycerol sebacic) acrylate.
PLA:	polylactic acid.
PLGA:	poly(lactic-co-glycolic acid).
POMaC:	Poly(octamethylene maleate (anhydride) citrate).
PPy:	PS: polystyrene.
PSBS:	poly(styrene-butadiene-styrene).
PVA:	poly(vinyl alcohol).
rGO:	reduced graphene oxide.
SCNT:	single-walled carbon nanotube.
SMD:	surface mounted devices.
TPC:	thermoplastic polyester elastomer.
UTS:	ultimate tensile strength.
VPS:	vapor phase soldering.

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