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Review

A review on recyclable printed electronics: Fabrication methods, inks, substrates, applications and environmental impacts

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Abstract: Innovations in industrial automation, information and communication technology (ICT), renewable energy, monitoring and sensing fields have been paving the way for *smart* devices, which can acquire and convey information to the internet, in every aspect of our lives. Since there is ever-increasing demand for large yet affordable production volumes for such devices, printed electronics has been attracting great attention in both industrial and academic research. In order to understand the potential and future prospects of the printed electronics, the present paper summarizes the basic principles and conventional approaches while providing the recent progresses in the fabrication and material technologies, applications and environmental impacts.

Keywords: Printed electronics, industrial automation, information and communication technologies (ICT), monitoring and sensing technologies, environmental impacts.

1. Introduction

Printed electronics has a great potential to offer biodegradable and recyclable solutions, which is a way forward to minimize the electronic waste (e-waste) caused by the ever-increasing number of disposable electronic devices [1,2]. In case of additive manufacturing (AM) of these devices, e.g. by conventional and the state-of-the-art printing methods, less material is used within few fabrication steps since the need for etching and masking is eliminated [3]. Hence, it is possible to improve the resource efficiency while reducing the fabrication costs since these manufacturing processes remain the same for both design prototyping and their mass production [4]. In addition, the recent advances also prove that AM methods can be used in hybrid printed electronic circuit fabrication, for which the conventional surface-mount technology (SMT) components are adhesively bonded to the printed substrates [5,6]. The full potential of using AM for electronic component manufacturing will be reached by replacing as many SMT components as possible with their printed counterparts as the assembly process may be completely eliminated [7–9]. Therefore, the electronic components and devices used in the fields of communication, energy and biomedicine can be viably manufactured using various printing technologies [10].

Printed electronics is a manufacturing process of registering thin conducting/semiconducting and dielectric material (*ink*) layer combinations on a low-cost *substrate* that may be recycled and/or naturally degrades in nature. Correspondingly, the manufacturing process is composed of three complementary stages: material selection, printing and post-printing. The materials for the printed electronics are principally inks of conducting, semiconducting or dielectric characteristics and substrates which are derived from synthetic or natural polymers. The inks are transferred with a master, i.e. through direct contact to the substrate, in case of contact printing while they are deposited onto the substrate typically through nozzles in case of non-contact printing. Unlike contact printing, non-contact printing methods are able to print digital models without need of printing unit/component changes. Posterior to the printing process, depending on the ink and substrate selection, it is often necessary to conduct sintering/curing in order to densify or crystallize the inks for the desired functional characteristics.

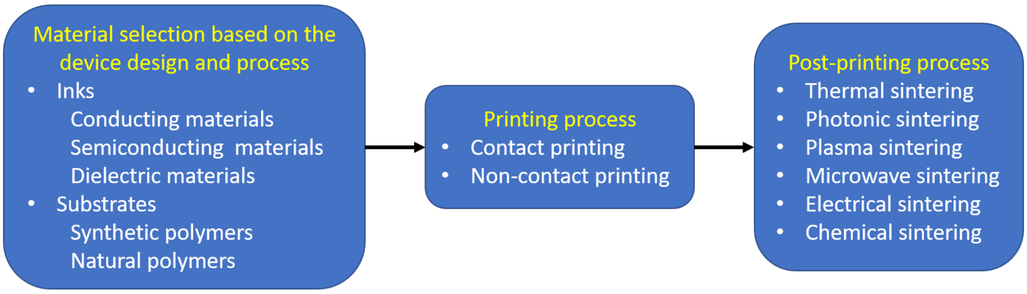

Figure 1. Complementary steps for electronics printing.

In Figure 1, these manufacturing stages are depicted. In these stages, inks, substrate and the printing technology must be carefully evaluated for quality, repeatability, and life-time aspects of the yield. Although the requirements for the mass production of printed electronics with the desired specifications are challenging, the current state-of-the-art for the materials, fabrication processes and inspection technologies demonstrate the increasing repeatability and device performance [11]. All these advances are closely associated. Undoubtedly, continuous growth of biodegradable and recyclable material portfolios for conductive, semiconductor and dielectric inks, and synthetic and natural polymeric substrates are fostered by the printing and post-treatment technologies, and optical inspection systems [12,13]. As a result, printing methods have gained ground for large area fabrication of flexible electronics including radio frequency identification (RFID) devices, photovoltaic cells, organic light-emitting diodes (OLEDs), thin film transistors (TFTs), diodes, displays, batteries and sensors detecting temperature, humidity, pH levels [14–17]. These developments demonstrate that reliable and repeatable solutions for the mass production of printed electronics with well-defined manufacturing standards will be available in very near future.

In the present study, we survey the printing principles and approaches while providing the recent investigations in the manufacturing and material technologies, applications and environmental impacts.

2. Motivation

The rapid growth of the Internet-of-Things (IoT) is contributing to the evergrowing interconnection of the digital world with the physical world. IoT contains a vast number of *smart* objects with communications capabilities for transferring data through wired or wireless communication networks. This includes, for example, various personal smart devices, as well as traffic management, security and RFID systems. The smart objects in general contain several sensing components (sensors) to acquire information about physical phenomena of interest and convey the data to the Internet to enable novel applications and services on the cloud. Naturally, IoT is a new frontier of information and communication technology (ICT), and sensor technologies.

IoT applications in healthcare, transportation and constructions, leisure and sports verticals enable transforming the objects from our everyday life into smart objects that can acquire and convey information to the Internet. This transformation inevitably requires addition of semiconductors, sensors and related electronics, which are designed to be discarded once the battery lifetime ends (possibly after a couple of thousands charge-discharge cycles). Although adding intelligence to the devices in everyday use is attractive and aids in digitalization of physical world, it also shortens their lifetime and becomes a major contributor to global e-waste [18]. The products that might have lasted 15 years must be hence replaced every 5 years after such transformation [19]. These periods become even shorter with the consumer intention towards purchasing

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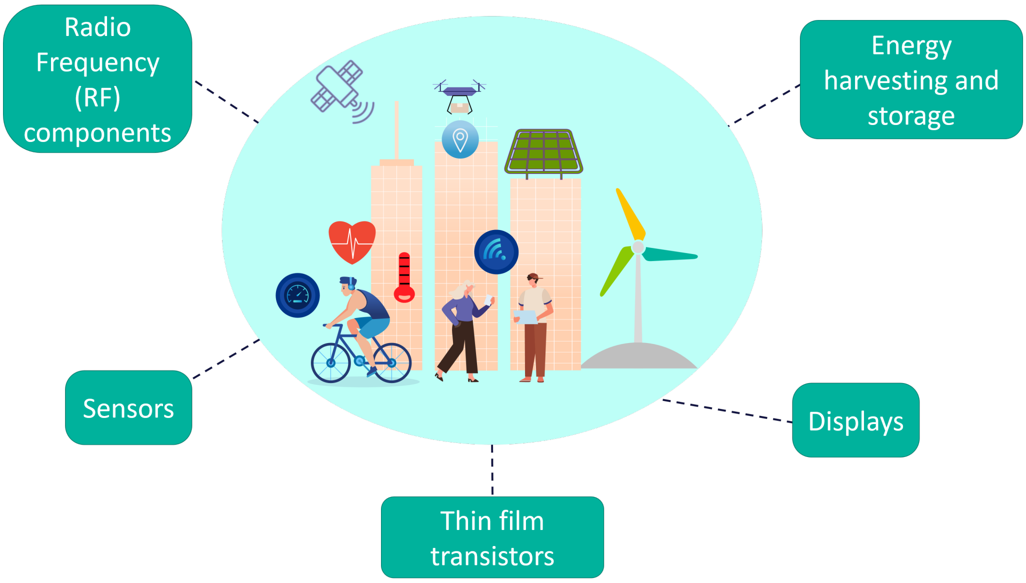

Figure 2. Printed electronic component classes reported in the present review.

new models of fully-operational electronic products. In addition, more power is needed as the use of the devices keep constantly increasing [20].

The consequences of the digital transformation and consumer behavior result in massive amounts of e-waste produced globally [21]. For instance, in 2019, approximately 53.6 Mt of e-waste was generated and it is increasing at an alarming rate of 2 Mt per year [22]. Out of these, only around 20-25% of e-waste are assumed to be formally recycled while the majority is domestically dumped into the environment or illegally exported to the developing countries [23,24]. Despite the claimed percentages of formal e-waste recycling, the absence of systematic procedures in the formal processes frequently ends up with the landfilling [25]. Improper handling of the matter, lack of environmental awareness and consumer behavior cause irreversible impacts on both environment and human health [26]. Therefore, the current trend of adding intelligence into everyday objects and discarding their environmental impact is not sustainable and requires alternative solutions that aims at reducing the looming e-waste stream.

Printed electronics offer several benefits, which are not likely to be achieved with the conventional electronics manufacturing. Kunnari et al. classified these benefits in terms of ecodesign, which are efficient use of materials, minimization of energy consumption both in the manufacturing and utilization phases, reduced use of hazardous substances, improvement of recyclability [27]. For instance, use of recycled and/or biobased materials, which reduces the use of raw/virgin materials, e.g. as printed electronics substrates, and recovery of silver and other precious metals used as the printing inks have been a long-term objective and already accomplished by the researchers in the field [28–32]. In addition to the substrate materials and their processing, the metal inks have been another issue in consideration to their high prices, resource depletion and ecological aspects (both in raw material mining and recycling) [33]. Therefore, separation techniques for metals from e-waste and development of energy-efficient and environmentally friendly carbon based or polymer inks have been investigated so as to replace or minimize the use of metal inks [34,35].

Printing technologies, as part of the additive manufacturing umbrella, have been extending electronics designers toolbox with access to various materials and ability to apply different structures in their designs. While printing of all the components is the ultimate aim of the electronics printing, most of the research activities are related to design and manufacturing of individual electronic components. Especially, their low manufacturing cost, large-area processability and lower carbon footprints compared

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with their conventional counterparts has attracted significant interest from the scientific and industrial communities. The printed electronic devices also provide favorable physical characteristics, e.g. low weight, stretching, resistance to folding or bending, which can not be realized with the conventional electronics [36,37].

As illustrated in Figure 2, researchers have reported passive, active and sensor printed electronic components [3,7]. While passive components are designed and manufactured based on electromagnetic properties of printed materials and shapes, active components utilize nonlinear behavior of printed materials, and sensors are designed based on the electro-mechanical features of the printed structures. A diverse range of active, passive and sensor devices including antennas, coils, RFID tags, strain gauges, gas, temperature and humidity sensors, color displays, OLEDs, photovoltaic cells, energy harvesters, TFTs, to name a few, have been printed on thin and flexible substrates which can be recyclable or even biodegradable [38–44]. In the following part of this section, recently investigated printed electronic component classes are summarized.

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2.1. Sensors

Table 1. Collection of substrate and ink types used in various printed sensors.

Sensor type	Substrate Type	Ink Type
Gas sensor [13]	-	Carbon nanotube (CNT)
Pressure sensor [13]	-	Carbon
Humidity sensor [45]	Polyethylene Terephthalate (PET)	Silver (Ag) nanoparticle & Poly(3,4-ethylenedioxythiophene) Polystyrene Sulfonate (PEDOT:PSS)
Humidity sensor [46]	Silicon/Silicon dioxide (SiO_2/Si)	Graphene PEDOT:PSS
Humidity sensor [47]	Paper	Ag nanoparticle
Humidity and temperature sensor [48]	Polyimide (PI)	Nafion/ Titanium monoxide (TiO)
		Nickel oxide (NiO)
Temperature sensor [45]	PET	Ag nanoparticle & PEDOT:PSS solution
Temperature sensor [47]	PI	Ag nanoparticle
		Carbon paste
		PEDOT:PSS solution
Temperature sensor [49]	Polyethylene (PE)	Silver nitrate ($AgNO_3$)
Temperature sensor [50]	Lead zirconate titanate (PZT)	Ag nanoparticle
Temperature sensor [51]	Paper	Ag nanoparticle
Temperature sensor [52]	PI	CNT PEDOT:PSS solution
Temperature sensor [53]	Glass	NiO
Temperature sensor [54]	Lithium niobate ($LiNbO_3$)	Ag nanoparticle
Temperature sensor [55]	Polyurethane (PU) plaster	Graphene PEDOT:PSS solution
Strain gauge sensor [56]	Buckypaper	Ag nanoparticle
Strain gauge sensor [57]	Polydimethylsiloxane (PDMS)	Ag nanoparticle & SBS/CB solution
		Carbon paste & SBS/CB solution

Sensors convert various physical phenomenon, e.g., acceleration, temperature, magnetic field and capacitance, into electrical signals. Consequently, sensors are vital components in a myriad of modern electronic applications across all industries. For instance, in manufacturing, for example, sensors can be employed to monitor ambient conditions and equipment parameters, to ensure the quality of the produced items. Moreover, various sensors are present in common household appliances, such as computers, security systems and smartphones.

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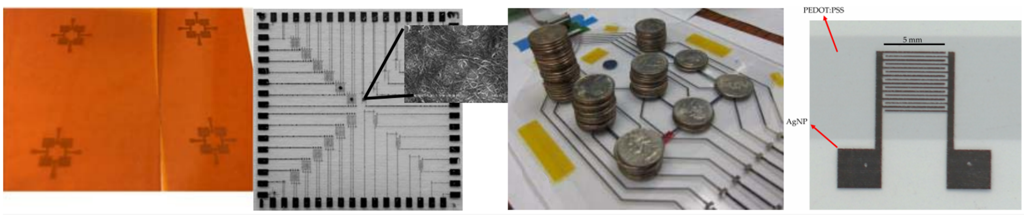
In printed electronics, it is common for a device to be partly printed and partly fabricated, as elaborated in the literature [58–60]. Various rigid components, such as memory units, sensors and chips that are manufactured through conventional methods can be combined with printed parts to form hybrid devices. However, with flexible and stretchable printed electronics, the use of these rigid components impose challenges in terms of mechanical and electrical integration. Although flexible interconnects and thinning of the microelectronic components can be used for this purpose [13], a more promising solution to this problem is to outright print these components. In particular, sensors can be manufactured using printing technologies, since flexible, biodegradable and stretchable sensing components compromise one of the most prominent area of printed electronics.

Various printed sensors are reported in the literature, e.g., carbon nanotube (CNT) based gas sensors [13], graphene-poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate) (PEDOT:PSS) humidity sensors [46], PEDOT:PSS based temperature sensors [45,47] and active matrix sensor arrays printed on flexible substrate comprising organic TFTs, organic photodiodes and printed devices [13]. Some of the printed sensors are depicted in Figure 3 while the substrates and ink types used are listed in Table 1. These studies also demonstrated the reasonable performance metrics in comparison with their conventionally manufactured counterparts.



(a) (b) (c) (d)

Figure 3. Printed sensors: (a) temperature sensors showing transparent property PEDOT:PSS (Reprinted with permission from [47]. Copyright 2020 IEEE); (b) a CNT gas sensor array; (c) a resistive pressure sensor array with test weights (Reprinted with permission from [13]. Copyright 2017 IEEE); (d) a PEDOT:PSS based temperature sensor with silver nanoparticle electrodes (Reprinted from [45] under CC BY 4.0 license).

Printed temperature sensors have been explored in detail by Rivadeneyra et al. [45] and Lall et al. [47]. The temperature sensors studied by Lall et al. were printed in a Wheatstone configuration using silver, carbon and PEDOT:PSS on a flexible polyimide (PI) substrate as presented in Figure 3(a). In addition, temperature sensor using this material combination was also presented by Rivadeneyra et al., the purpose of which was to generate a combination of both positive temperature coefficient (PTC) and negative temperature coefficient (NTC) materials and hence to increase sensitivity and measure a broad range of temperatures. As seen in Figure 4, the resistance of PTC materials increases as temperature rises, vice versa, the resistance of NTC materials decreases when temperature increases. Silver functions as a PTC material while carbon and PEDOT:PSS exhibit NTC behaviour. Rivadeneyra et al. [45] also studied specifically PEDOT:PSS based temperature sensors printed on a flexible polyethylene terephthalate (PET) substrate (see Figure 3 (d)), and the effects of electrode spacing and fabrication methods on the thermal sensitivity of the sensors.

The investigations by Lall et al. demonstrated that temperature sensors printed using silver ink can reach to very high sensitivity values achieving 0.192 %/°C temperature coefficient of resistance (TCR) [47]. Furthermore, in the measured temperature range, resistivity of silver is reported to behave in a highly linear fashion, as illustrated in Figure 4. In contrast, the TCR value of carbon is -0.048 %/°C and -0.051 %/°C with PEDOT:PSS. In addition, the NTC of these materials changes non-linearly for some

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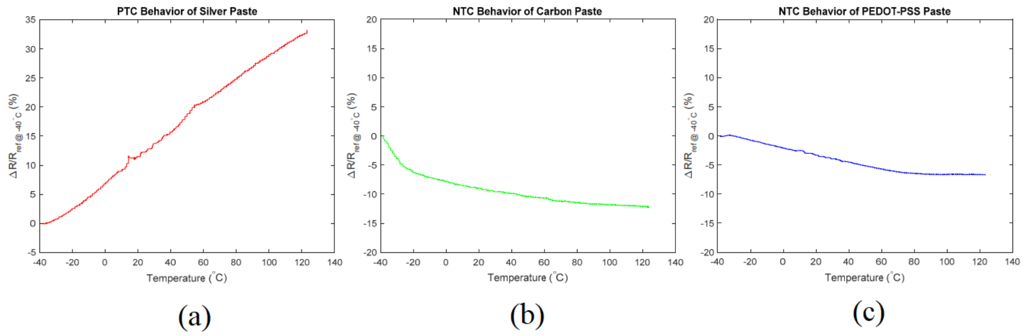

Figure 4. (a) Highly linear PTC behaviour of silver throughout the test temperature range; (b) NTC behaviour of carbon; (c) NTC behaviour of PEDOT:PSS. Note that carbon exhibits near linear NTC behaviour after an initial region of non-linear behaviour (roughly from -40°C to -20°C). On the other hand, PEDOT:PSS behaves near linearly initially from around -40°C to 70°C. (Reprinted with permission from [47]. Copyright 2020 IEEE)

temperature ranges. The PTC and NTC characteristics of just mentioned materials are given in Figure 4.

In the work by Rivadeneyra et al., it was concluded that the sensitivity of printed temperature sensors could be increased by altering the order of fabrication steps [45]. Higher sensitivity values were obtained by following a sequence of first printing and drying the silver electrodes, then printing and drying PEDOT:PSS and finally, sintering the sensor. In addition, sensitivity of these sensor modules were also enhanced by a factor of 2.2 as a result of increase in electrode spacing from 150 μm to 200 μm.

Strain gauge sensors, as being another common sensor application, are used across numerous industries for monitoring strains caused by external forces and/or moments. As seen in Figure 5, such sensors are used to monitor curing and exerted pressure during the manufacturing process, and structural integrity and damage during the operation, i.e. structural health monitoring (SHM) [61,62]. Recently, aerosol-jet printed CNT strain gauge sensors with CNT onto buckypaper embedded in composite laminates demonstrated such sensor implementations in both resin flow and curing monitoring [56]. Thus, the effects of process defects can be simply monitored during the entire life span of the structure, which gives a clear idea about possible structural failures in advance. In a more recent paper by Wang et al., high sensitivity/low hysteresis screen printed strain sensors are also demonstrated. The presented sensors have good performance metrics comparable to their commercial counterparts [57]. This further indicates the great potential for printability of strain sensors.

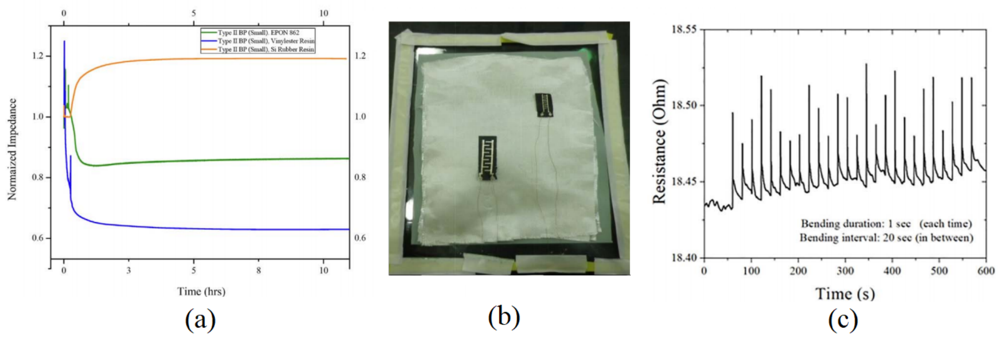

Figure 5. (a) Normalized impedance over time for vacuum and curing cycles for three types of resin. (b) Printed strain sensor placed on glass fiber reinforced composite. (c) Results of impact hammer bending of glass fiber reinforced composite with embedded printed strain sensor. (Reprinted from [56] under CC BY-NC-ND 4.0 license)

Furthermore, printed organic transistors can be also used for sensing slowly changing physical phenomenon. For instance, a bacteria sensor, in which bacteria binding electrodes are integrated with PEDOT:PSS transistor, was demonstrated by Demuru et al [63]. Similar low frequency sensors for measuring arterial pulse waves were reported by Laurila et al. [64], for which the sensor was printed from piezoelectric poly(vinylidenefluoride-co-trifluoroethylene) (PVDF-TrFE) and accompanied with entirely printed amplifier.

2.2. Thin film transistors and their applications

Digital electronics rely on the nonlinear behavior of diodes and transistors. However, printed transistors are known to be relatively large and requiring high supply voltage. Recently, carbon based transistors have been developed to overcome these issues. For example, Portilla et al. and Williams et al. recently demonstrated low power printed TFTs, which is a step forward to fully recyclable electronics [65–67].

In addition to low power TFTs, multiple transistors have been recently combined with larger electronic circuits by Matsui et al. [68] and Sun et al. [69] to develop operational amplifiers. In their investigations, Matsui et al. used n-type semiconductor from 8-bis[5-(3-cyanophenyl)thiophene-2-yl] benzo[1,2-c:4,5-c']bis[1,2,5] thiadiazole derivative and p-type semiconductor from 2, 8-difluoro-5, 11-bis(triethylsilylethynyl) anthradithiophene (diF-TES-ADT). On the other hand, Sun et al. used pentacene as the semiconducting material. Furthermore, printing technologies, especially inkjet printing, have been observed to be suitable for fabricating latches by Weller et al. [70], AND and OR circuits by Kamali-Sarvestani et al. [71], and inverters by Singh et al. [72]. In the investigations by Weller et al., latch circuits were implemented by using PEDOT:PSS transistors while AND and OR circuits were printed by using single-walled CNT field effect transistors (SWCNT-FET) in the research study of Kamali-Sarvestani et al. The inverters, on the other hand, were printed using dithiophene (DTBDT-C6) and polystyrene (PS) semiconducting material as part of the investigations carried out by Singh et al.

Integration of printed transistors with sensors, antennas and active circuits has been widely investigated in the literature. The current printed transistor technologies cover applications with relatively modest frequency needs, e.g. in sensors, RFID and audio applications. For instance, an interesting audio application was presented by Kheradmand-Boroujeni et al. [73], for which the printed preamplifier was combined with printed piezoelectric loudspeaker. The amplifier was constructed from organic FETs (OFETs), printed capacitors and resistors while the whole circuit was printed on recyclable PET sheet.

2.3. Radio Frequency (RF) components

RFID is a technology that automates the process of extracting data from RFID tags or smart labels and identifying them [74,75]. As the name suggests, RFID is used to transfer data wirelessly via RF transmissions. As schematized in Figure 6, a typical RFID system consists of three parts: RFID reader, RFID tag or smart label categorized as active or passive, and a computer that collects and manages the information [75]. In numerous RFID applications, the computer manages a database based on the received information. In passive RFID tags, no power supply units are integrated into the circuits [76]. Instead, these tags draw power wirelessly through induction from the device that is reading them. Active tags, on the other hand, have an onboard power supply. However, passive tags are more common than active tags due to them being more compact and less expensive [74].

In RF electronic hardware designs, power losses incurred in components and interconnects need to be minimized, as these losses directly define the quality of the end-product. At mm-wave frequency, inkjet printed interconnects have been found to outperform conventional RF ribbon-bond interconnects, as reported by Eid et al. [77] and presented in Figure 7 (a). The printed interconnects incur less losses at mm-wave frequency and generally are closer to a continuous transmission line than the ribbon-

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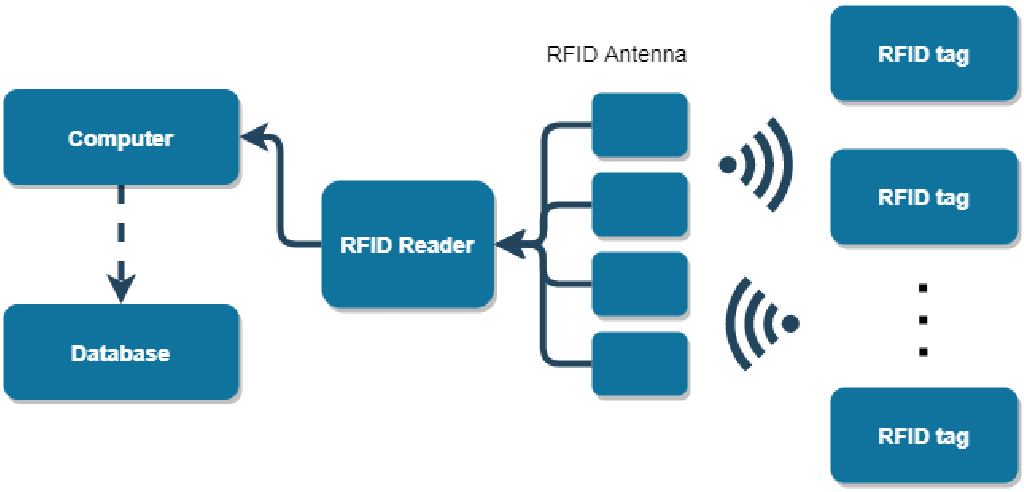
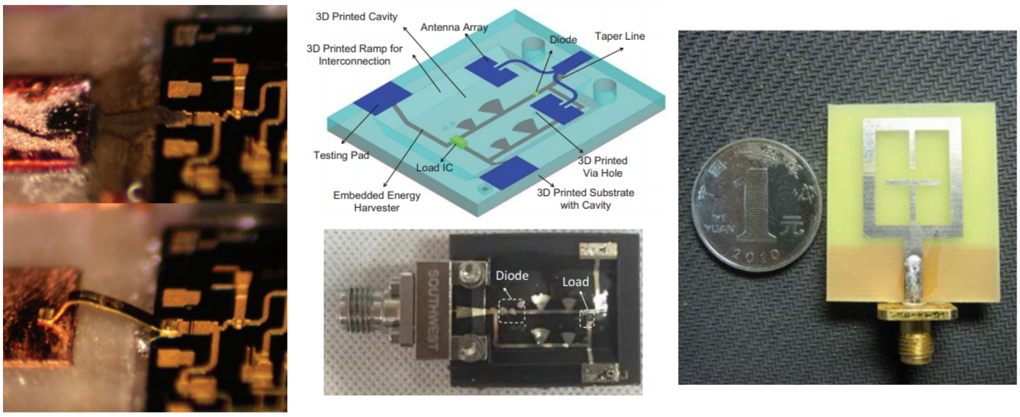


Figure 6. An illustration of an RFID system consisting of multiple RFID tags and an RFID reader connected to a computer managing a database.



(a) (b) (c)

Figure 7. (a) Above is a printed interconnect and below is a ribbon-bond interconnect (Reprinted with permission from [77]. Copyright 2020 IEEE). (b) Above is the structure for the embedded-on-package 5G energy harvester and below is the printed device (Reprinted with permission from [78]. Copyright 2019 IEEE). (c) Printed antenna for WLAN and 5G applications (Reprinted with permission from [79]. Copyright 2020 IEEE).

bond interconnects. Furthermore, a printed antenna applicable to 3.5 GHz 5G, as well as wireless local area networks (WLAN), was demonstrated by Wang et al. [79], which is shown in Figure 7 (c). In addition to this, Jilani et al. [80] also presented an inkjet printed antenna for mm-wave 5G, which was able to function in the 28 and 38 GHz frequency bands.

Nowadays, owing to the printing technologies, RFID tags can be manufactured in highly cost efficient manner. This opens up the possibility of using RFID tags, for example, as part of packaging and other applications (please, see Figure 8), for which the device has short life expectancy and is ultimately disposed. Literature on this subject has shown that manufacturing of RFID tags is not limited to a specific printing technology [58,59,81]. For instance, gravure, screen, flexographic and inkjet printing technologies have been effectively used in RF component manufacturing.

Printed RFID tags are able to function in the same frequency bands as once produced through conventional methods. This includes both the high frequency (HF) band and the ultra high frequency (UHF) band of 858-930 MHz, which enables the simultaneous detection of multiple RFID tags [59]. Typically, printed HF RFID devices operate at 13.56

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MHz [58,82], while on the other hand, printed UHF RFID devices typically operate at 868 MHz [58–60]. A possible explanation for the specific use of 13.56 MHz frequency for HF RFID seems to be the commitment to the SFS-ISO/IEC 18000-3:en standard [83]. This standard describes parameters for air interface communications at 13.56 MHz for RFID item management. Similarly for UHF RFID, ETSI EN 308 208 standard [84] enables 2 W RFID transponders operating in the 865-868 MHz frequency band, which has motivated the printed UHF RFID tags [58–60].

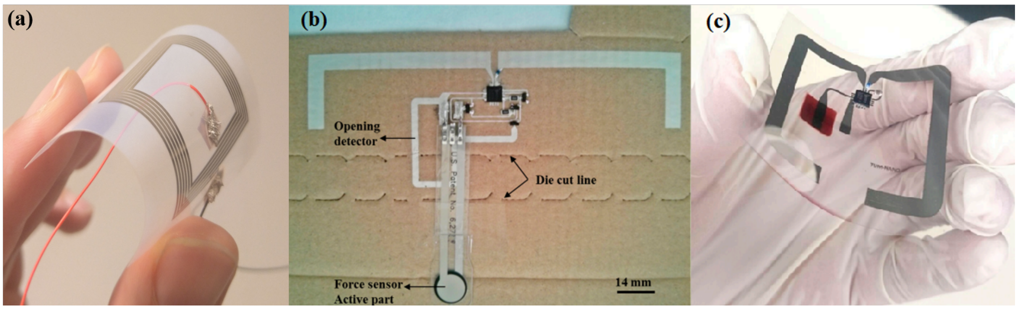
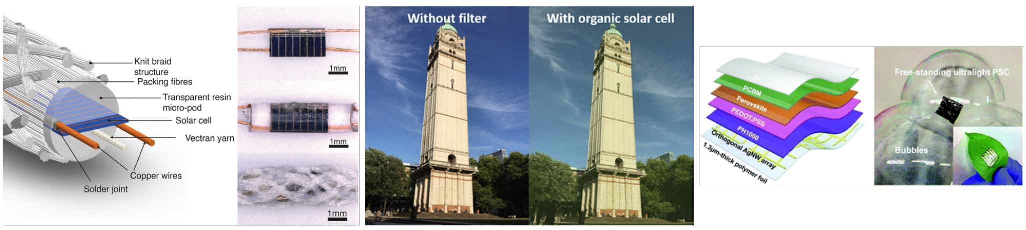


Figure 8. (a) A printed RFID coil by the authors; (b) a fabricated RFID tag with an openning sensor and force sensor as a part of packaging [59]; (c) an RFID tag with light detection capabilities printed on a flexible PEN substrate [60]

2.4. Energy harvesting and storage

Energy harvesting modules, as exemplified in Figure 7 (b), have been gaining interest for use in low-power and low-cost IoT devices. Recently, Lin et al. [78] presented a package-integrated mm-wave 5G energy harvester fabricated with 3D-printing and inkjet printing technologies. As discussed by Eid et al. [77], these system-on-package energy harvesters could be also designed to harvest other forms of energy, such as solar and vibration energy. The capability of harvesting energy from the environment would allow greater autonomy of various small IoT devices compared to their battery powered variants.



(a) (b) (c)

Figure 9. (a) Solar cell embedded into yarns (Reprinted from [85] under CC BY 4.0 license), (b) Transparent organic solar cell (Reprinted with permission from [86]. Copyright 2020 American Chemical Society), (c) Flexible perovskite solar cell (Reprinted with permission [87]. Copyright 2019 Royal Society of Chemistry).

Solar cells use photoelectric effect for converting light energy to electricity. Simple solar cells are composed of supporting layer covered by photovoltaic or photoemissive cells and conductive transport grid. The cells convert light to electricity that is carried away by transport layer on top of the cells.

The most popular semiconducting material for solar cells has been silicon, the cells out of which have an electricity conversion factor over 20 %. However, silicon is not recyclable and thus, not an environmentally friendly option. On the other hand, printable and recyclable solar cells can be created from perovskite or other organic compounds [88,89]. For instance, with the reported efficiency of 23 % by Kim et al., perovskite solar cells are approaching efficiency of silicon cells [90]. Besides, organic

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solar cells have been also gaining attention due to their environmental friendliness [91]. However, their wide scale use is hampered by low power conversion efficiency because of high recombination loss that leads to high voltage loss [92]. This problem has been overcome with the use of new materials by Liu et al. [93]; hence, printed organic cells have reached 15 % of power conversion efficiency. As a result, organic cells are becoming an attractive option for fully printed electronics.

Industrial scale manufacturing of printed and thin organic solar cells is extremely cost efficient, and it allows manufacturing of flexible, lightweight and transparent cells [94]. In addition, elastic solar cells have been manufactured and embedded into textiles, which paves a way for solar energy harvesting cloths. For instance, woven solar cells were demonstrated to generate over 2 mW/cm^2 by Satharasinghe et al. [85], which was sufficient to power a mobile phone. Furthermore, recent investigations by Kang et al. [87] and Lee et al. [86] showed that organic solar cells could be fabricated extremely thin, lightweight and transparent, which simply finds applications in optoelectronics devices.

2.5. Displays

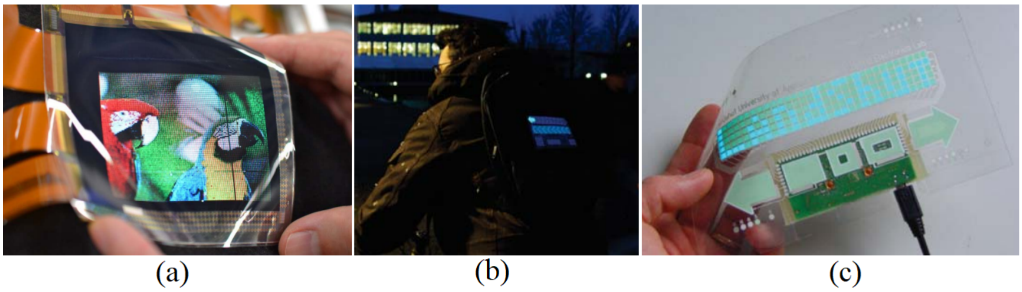

Figure 10. (a) A flexible color display (Reprinted with permission from [95]. Copyright 2018 IEEE), (b) a printed electroluminescent matrix display integrated into a back pack, (c) a closer image of the previously mentioned screen printed matrix display with the controlling module (Reprinted with permission from [96]. Copyright 2018 IEEE).

Printed organic transistors are attractive due to their flexibility, low manufacturing cost and possibility to cover large structures or sheets cost efficiently. Those features have fueled the success story of active matrix OLED (AMOLED) displays. Usage of organic transistors in displays have been hindered by their low electron mobility. However, recently, Mizukami et al. [95] demonstrated how dithiophene based organic TFTs (OTFTs) can be used as a light source in printed displays. In their work, flexible color displays were fabricated with inkjet printing technology (please, see Figure 10 (a). As depicted in Figures 10 (b) and (c), a novel study on printed displays have been reported by Ivanov [96], in which a screen printed electroluminescent matrix display were developed and described in detail.

3. Printing technologies

The technologies used for printing electronic components are well-known in the graphic arts, and some example options are gravure printing, flexography, offset printing, screen printing, and inkjet printing as depicted in Figure 11. These technologies are often divided in contact printing methods, which use printing master [97], contactless (non impact) methods that do not need a master [98], and in hybrid printing processes combining different printing and deposition techniques. Hybrid printing [99] is used to deposit stacked layer structures, such as electrodes, conductive layers, isolating layers for electronic components. After deposition, the ink subsequently changes its phase in the desired site [100–103]. The initial state, flow and phase change of the ink on the substrate are mainly affected by the viscosity, density, surface tension, solvent evaporation rate, solubility and curing characteristics of the ink, and wettability and permeability of the substrate in use [104,105]. Moreover, multi-layer and multi-material nature of the final device require compatibility and hence treatment of each layer to ensure the desired layer function, e.g. minimal sheet resistance, within a short-time span and without damaging previously deposited layers. As exemplified in the previous section, typical printed devices include sensors, batteries, capacitors, transistors, solar cells, memories, electroluminescent structures, large screens, and light panels [34,106–115].

A well-known application of printed electronics is RFID systems where the antennas can be printed. Printing the RFID antennas on flexible substrates is a very efficient solution because it provides thinner, lighter and cheaper structures compared to the conventional etching-based manufactured antennas. Gravure printing, offset and flexographic printing methods are more commonly used for high-volume production needs, such as solar cells. Offset and flexographic printing are primarily used for inorganic and organic conductors. Gravure printing is suitable for high resolution and quality applications and layers. Such are e.g. organic semiconductors and semiconductor/dielectric interfaces of transistor structures. There have been hybrid processes under investigation, where different printing and deposition techniques, such as gravure printing, via-hole screen printing and electroless plating are combined [116,117].

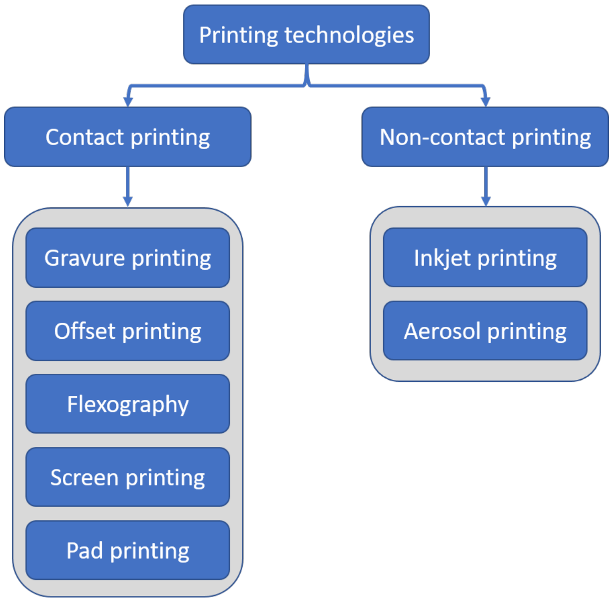

Figure 11. Classification of printing technologies.

The key properties and parameters of the different printing techniques are summarized in Table 2. Flexographic printing creates a thin printed layer with a feature size of 80 μm and a throughput of 3-30 m^2/s. Offset and gravure printing provide similar features, the latter being the fastest printing technology in terms of print press speed. Screen printing and inkjet printing can give a larger layer thickness, inkjet printing up to 20 μm and screen printing up to 100 μm, even with a lower throughput of 2-3 m^2/s in screen printing and 0.01-0.5 m^2/s in inkjet printing. Inkjet printing has traditionally been better suited for R&D or special applications, screen printing is excellent for stacking multiple thick prints, while rotogravure and flexographic printing offer more opportunities for mass production of printed electronics [118].

Table 2. Key properties and parameters of different printing methods.

	Gravure	Offset	Flexography	Screen	Inkjet
Throughput (m^2/s)	3-60	3-30	3-30	2-3	0.01-0.5
Resolution (lines/cm)	20-400	100-200	60	50	60-250
Printing speed (m/min)	100-1000	100-900	100-700	10-15	15-500

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3.1. Contact printing methods with a master

In contact printing techniques, ink is transferred through direct contact between the ink and substrate. Contact printing is often referred to as transfer printing or roll-to-roll (R2R) printing. These printing techniques typically utilize a roll to transfer ink directly to the substrate. The initial costs are high and these techniques require longer preparation periods [102]. However, lower production costs, high manufacturing speeds and the repeatability associated with contact printing technologies make them favorable for mass production [12].

3.1.1. Gravure printing

In gravure printing, the elements of the image are engraved on the surface of the printing cylinder, while the non-image areas remain at the original level. The engraved printing cylinder rotates in an ink fountain partially filled with ink. The entire surface of the cylinder collects ink from the fountain and the excess ink is wiped off the non-image areas with a doctor blade leaving the ink in the engraved cells. The ink is then transferred from the cells directly to the substrate in a printing nip under pressure. Ink transfer is often improved by using an electrostatic assistance system (ESA) that creates an electric field across the nip. The electric field assist in lifting ink from the cells onto the substrate. The ink layer is dried by evaporating the ink solvent with hot air [97,119].

Gravure printing is especially suitable for long printing times, because the production of a printing cylinder is time consuming and expensive [97,119]. As a result, gravure printing has lost its market share recently due to the trend towards shorter editions and more individualized prints, whereas gravure printing run lengths typically range from hundreds of ashes to over a million copies. Gravure printing is known for its high print quality and speed. For example, the resolution is 20 to 400 lines/cm and the print speed is 13 to 16 m/s. Other benefits include a simple printing process, accurate ink application, and flexibility in press design. Publication presses are designed for fast printing of high-quality magazines, catalogs and brochures. Print quality has further improved when laser engraving came more popular. However, increased demand for short batches and personalization has reduced the market share of rotogravure printing.

3.1.2. Offset printing

Offset printing is an indirect printing method in which ink is transferred from a printing plate to a substrate with an intermediate blanket cylinder. Surface chemistry has an important role in offset printing. Damping rollers apply a thin layer of moisturizing water to the hydrophilic non-image areas of the plate. After that ink rollers transfer a thin film of ink over the oleophilic image areas of the plate. The image and other areas of the plate are at the same level, but their surface energies are different. The image areas accept ink but repel water and non-image areas accept water but repel ink. This difference in surface energy prevents ink from spreading from the image areas, and a thin layer of wetting water in non-image areas prevents further spreading. The ink layer is then transferred to the blanket cylinder and further to the paper in the nip under pressure. Offset printing is a wet-on-wet printing method in which process inks are printed sequentially without intermediate drying. Therefore, some ink is also transferred to the blanket cylinder of the next print unit. After the last printing, the ink layer is dried by absorption, polymerization, oxidation, or evaporation [119].

3.1.3. Flexography

Flexography uses soft and flexible printing plates in which the pixels are raised on the top surface of the rest of the printing plate. The printing ink is applied to the pixels through an anilox roll with small cells evenly engraved on the surface. The surface of the anilox roll is applied with ink from the chamber and the excess ink is then removed with a scraper blade. After formulation, the ink is located only in the cells of the anilox roll. The ink is then transferred to the pixels on the surface of the printing plate. The

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ink transfer takes place in a nip where the prevailing pressure allows the ink transfer by improving the contact between the ink and the substrate. At the nip outlet, the ink layer splits and some of the ink is transferred to the substrate. The ink layer is dried by blowing hot air onto its surface. This causes the ink solvent to evaporate.

Flexography is used e.g. for printing RFID antennas, batteries, organic electronic circuits such as OLEDs and solar cells. OLEDs are used to make diode displays (televisions, computer monitors) and lighting (diode lamps). Smart labels and thin films printed on polyester film can be used, for example, to monitor temperature during drug transport [120,121]. The resolution is typically 60 lines/cm and the print speed is 3 to 12 m/s. The biggest challenge is maintaining color saturation and density in fixed areas. Flexography is suitable for a wide range of printing media. For example, non-porous and porous substrates can be printed without problems.

3.1.4. Screen printing

Screen printing is a so-called push-through process in which ink is pushed through a fine fabric (screen) made of plastic or metal fiber or wire. The non-image areas of the screen are covered with a stencil that defines the printed image and acts as a printing plate. The screen is full of ink that is pushed through the image areas of the screen using a squeegee. The achievable print thickness is greater than in other printing methods. Drying of the ink layer is typically accomplished by evaporation, oxidation, or ultraviolet (UV) curing. Drying can be accelerated by blowing warm air onto its surface [119]. Print quality is determined by the material, fineness and thickness of the screen, the distance between the top and bottom of the screen, and the open area of the screen. The fineness of the screen is typically 90 to 120 fibers/cm.

Screen printing is a versatile and simple process for transferring ink to the surface of substrates. The range of substrates is wide, from paper to ceramics in both sheets and continuous paths. The technology also allows printing on curved surfaces. Applications range from art to large industrial applications, from small electrical components to multi-square meter commercials, and from single-piece production to long print runs. Screen printing is limited by relatively modest print quality. Screen printing is the oldest technology used to print electronics. In addition, it is the cheapest, simplest and the most flexible printing technology. It can be used e.g. for printing electromagnetic enclosures, capacitors, membrane switches or transistor electrodes. Screen printing has also been studied to print CNTs and panels of graphene nanoparticles on polymer substrates [122,123] and flexible printed supercapacitor structures comprising aqueous electrolyte and carbon electrode [117].

3.1.5. Pad printing

Pad printing is a printing process that can be used to transfer a planar image to a 3D object. This is accomplished using an indirect offset printing technique in which an image is transferred from a cliché through a silicone pad to a substrate. Pad printing has been used as an alternative to screen printing, e.g. for printing transistor electrodes [124].

3.2. Non-contact printing methods

In non-contact printing, nozzles are typically used to accurately deposit inks onto substrates without direct contact, which in turn reduces nozzle contamination. Non-contact printing technologies are significantly slower than R2R contact printing methods. However, many non-contact printing methods are able to print digital models, unlike contact printing methods that require parts of the printing unit to be changed in order to print various patterns. This is particularly efficient in prototyping where a model may be printed only once, and for on-demand production [102].

3.2.1. Inkjet printing

An inkjet is a digital printing method, in which liquid ink droplets are sprayed from small nozzles directly onto the surface of the paper according to digital page information. Nozzle groups are located at the print heads, which are either stationary and page-wide or smaller elements that scan from page to page. Fixed nozzle groups can be up to 1 m wide, allowing for higher print speeds [125]. Inkjet printing is a fairly simple printing method and also has a light and compact structure. In addition, each printed page can be different and no printing plates are required, allowing for personalization and on-demand printing. Therefore, the inkjet is suitable for hybrid printing applications used in conjunction with the traditional printing techniques.

The main advantage of inkjet printing is high print quality (resolution 2880 dpi), but limited print speed (2 to 5 m/s) [125]. The inkjet is widely used in commercial, graphic and packaging printing as well as in publication printing. For example, direct mail and wide-format products are often printed with an inkjet. In addition, the versatility of the inkjet process for both sheets and web printing increases its use. The use of UV inks has grown strongly because they are suitable for use with a variety of substrates, do not produce volatile organic compounds, and are more durable [126].

Inkjet technologies can be divided into continuous-stream (CS) and drop-on-demand (DOD) inkjet. These techniques are further subdivided into sub-technologies that differ in droplet formation or control mechanisms [125]. CS is mainly used in high-speed and high-volume printing applications such as direct marking, bar coding, personalization, labeling, catalogs, hybrid printing, packaging and direct mail. Typically, there are no speed limits, but print quality depends on droplet size and distance, nozzle diameter, droplet stream quality and continuity, and ink surface tension and viscosity. New developments have increased the resolution of the continuous inkjet printer to 600 dpi. A continuous inkjet generates a continuous stream of ink droplets, but only some of them are printed on the substrate. The ink droplets are charged according to the digital page information, and the charged unprinted droplets are then directed to the gutter and collected to the ink chamber. The ink drop is recycled about 40 times before printing [125]. On the other hand, the high-resolution DOD inkjet is primarily used for small-scale home and office printing, desktop publishing, and high-quality wide-format printing and color coating. However, the print speed is slow. The maximum resolution (2880 dpi) is up to five times higher than CS inkjet, and the droplet size is also significantly smaller. In the DOD inkjet, every drop of ink hits the paper, and no charging, deflection or recycling systems are required. The most common DOD inkjet technologies are thermal and piezoelectric inkjets, which differ in formation technology.

3.2.2. Aerosol jet printing

Aerosol jet printing, also known as Maskless Mesoscale Materials Deposition (M3D), utilizes atomization of an ink, which produces very small droplets in the aerosol ranging in size from 1 to 5 μm in diameter [127]. These droplets are delivered to the ceramic nozzle attached to the print head with a vacuum generated by means of a nitrogen sheath gas stream and impinged as a high-velocity jet onto the surface of the substrate [128]. This process enables printing on both plane and conformal surfaces. In order to print complex and 3D patterns, it is crucial to have accurate control over the beam. Thus, a shutter in front of the nozzle is used to selectively interrupt the beam. In addition, a proper distance between the nozzle and substrate, which should not exceed 10 mm and should not be lower than 1 mm, must be also maintained for the accuracy. Exceeding these boundaries usually cause overspray defects in the printed pattern.

Similarly to inkjet printing, aerosol-jet printing is capable of printing designs based on digital models. As an advantage, aerosol-jet printing does not suffer from nozzle clogging. Feature sizes as small as 10 μm can be produced with this technology, allowing high resolution printing [129]. However, the main drawback of the aerosol-jet printing is the speed, which is up to 12 m/min; thus, it is not suitable for mass manufacturing of printed electronics [102].

4. Printed electronic materials

4.1. Inks for printed electronics

In printed electronics, inks are used in order to carry out a certain function. In order to print more complex electronic structures several kinds of inks are required, such as conducting, semiconducting, dielectric (or insulator inks). In some specific cases, light-emitting or photovoltaic inks are also used [130]. The functional inks used in printed electronics need to be able to form homogeneous layers and should be compatible with the other inks that are simultaneously applied. Functional inks usually contain solvents, resins and/or polymers. Instead of pigments, metal particles are commonly used and additives, like dispersants, can be also utilized in order to modify the properties of the ink based on the application [115]. The components in inks can be both organic and inorganic materials; however, some materials have to be in micro- or nanoscale in order to minimize the printer clogging issues [131].

4.1.1. Conducting materials

The conducting component in the ink can be a metal, ceramic or polymer. The ink structure can be in the form of dispersed nanoparticles, dissolved organometallic compounds, dissolved or dispersed conductive polymers [39]. Nevertheless, metals are the most popular conductive materials due to their comparable conductivity. There are mainly two kinds of metal-based conductive inks, which are nanoparticle suspensions and metal-organic decomposition (MOD). Metal-nanoparticles are easy to disperse into inks for different printing methods due to their nanometer size; however, the production of metal-nanoparticles require high labor and energy input. Additionally, stabilizers are required to prevent agglomeration in the inks and the post-print treatments require high temperatures (simply over 100°), but can be reduced by decreasing the size of the nanoparticles [115,120]. MOD inks are based on metal precursor dissolved in a compatible solvent; thus, there are no particles left without agglomeration or condensation problems. The evaporation of the solvent is simple and inexpensive; however, there is a volume loss of 80 %, resulting in gaps in the printed conductive pattern. Decrease in conductivity is inevitable; nevertheless, the conductivity can be increased by printing multiple layers with such inks [131,132].

The most popular metal inks are silver, copper, gold and aluminium, for which silver and aluminium have the highest and lowest conductivities, respectively. Silver-based inks have high conductivity and resistance to oxidation for long periods of time but silver is an expensive metal. Compared to silver-based inks, copper-based inks have only 6 % less conductivity. It is considerably affordable; however, the conductivity decreases over time due to oxidation and high sintering temperatures are required. There are several methods to prevent the oxidation of copper, such as use of antioxidants or carrying out the synthesis in an organic solvent and forming a protective layer. These methods are, nonetheless, short-term solutions while long-term solutions against oxidation are e.g. forming a dense shell of non-oxidizable conductive material or a biometallic core-shell [39]. In addition to these commonly used metals, gold is another easy to prepare metal as an ink, environmentally stable and requires relatively low sintering temperatures in order to function as a conductor; however, the price of gold is high. Aluminium has a high tendency for oxidation and is more chemically active compared to copper. CNT and graphene are the most interesting carbon-based conductive inks; however, carbon fibers and particles can also be used for conductive inks. There are two types of CNTs, metallic and semiconductive, these can be separated, even if it is challenging, and used in different inks. Metallic SWCNT (m-SWCNT) have excellent stability, flexibility, light transmittance and its conductivity increases with the thickness. Graphene has a high conductivity, light transmittance, mechanical strength and elasticity;

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however, conductivity increases and light transmittance decreases with an increasing amount of layers [115,120,131].

Conductive oxide ceramics are usually produced by doping in order to alter the cation or the anion lattice. Examples of doped conductive oxide ceramics are antimony tin oxide (ATO), fluorin tin oxide (FTO), indium tin oxide (ITO), aluminium zinc oxide (AZO) and gallium zinc oxide (GZO). ITO is the most used of these materials, because of its superior conductivity; however, it is a rare and expensive material [131]. There are two types of ITO inks, sol-gel and nanoparticle, the sol-gel ink has better conductivity, but requires very high sintering temperatures, while the nanoparticle ink cannot form dense oxide films with high conductivity [120].

In general, conductive polymer materials have lower electrical conductivity than metallic materials. Conductive polymers are often e.g. flexible, soft, lightweight, cheap and compatible with aqueous and organic solvents. The production of conductive polymer inks is challenging, due to their limited solubility, stability and processability. The concentration of polymer inks is very low, 1-6 %, which results in long drying times and thin films [115]. Conductive polymers can be conjugated polymers, polymer electrolytes, organic metal chelates or charge transfer complexes. The conductive polymer with the highest conductivity and best temperature and humidity stability is PEDOT:PSS. PEDOT is insoluble, but it can be made soluble by using PSS as a dispersion. PEDOT:PSS has a high light transmittance, stability, flexibility, moderate band gap and low redox potential. The conductivity of PEDOT:PSS can be further improved by adding organic compounds or post-treatment using various compounds. Other conductive polymers that have been used in inks are polyacetylene, polyaniline, polypyrrole, polyacene, polythiophene, polyparaphenylene, polypyrrole and doped polyacetylene [120,131].

4.1.2. Semiconducting materials

The semiconducting layer of the printed electronics, serves as the active layer, where most of the electric activity occurs. Silicon and germanium have been the most popular semiconductor materials; however they are neither recyclable nor biodegradable. Both CNTs and graphene can be used as semiconductors [120]. Semiconducting SWCNT (s-SWCNT) has high flexibility, light transmittance and mobility. By using only 1-2 layers of graphene it can be used as a semiconductor [115]. Many ceramic oxides can be used as semiconductor materials, such as tin oxide (SnO_2), zinc oxide (ZnO), indium oxide (In_2O_3) and gallium oxide (Ga_2O_3) [131,133]. The ceramic oxides have non-toxic degradation; however, they require high sintering temperatures and are expensive due to their rarity [1].

Polymer semiconductors can generally be divided into two group, p-type and n-type polymer semiconductors. Polymers mostly using holes as carriers are p-type polymer semiconductors, the most promising are polythiophenes (PT) and polyfluorenes (PF). Examples of PTs are poly(3-alkylthiophene) (P3AT), poly(3-hexaylthiophene) (P3HT) and poly(3, 3'-dialkyltetrathiophene) (PQT). N-type polymer conductors use electrons as carriers, like poly(9,9-dioctylfluorene-co-bithiophene) (F8T2) [131].

4.1.3. Dielectric materials

Dielectric inks are used as insulator and capacitor layers in printed electronics. Nonetheless, it is challenging to make and print dielectric inks, compared to conducting or semiconducting inks. The dielectric layer requires enough thickness in order to prevent electric leakage. Substrate materials, like cellulose, gelatine, shellac and silk, are insulators, and can be used as dielectrics [1]. Additionally, ceramic oxides can be used as dielectric materials; however, they have a tendency to form pinholes and cracks [131].

There is an abundance of polymers suitable as dielectrics, which have low surface roughness, surface trap density, concentration of impurities, cost and sintering temperatures. Additionally, they are also compatible with organic semiconductors. The most commonly used polymer dielectric materials are polymethyl methacrylate (PMMA),

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PI, polyvinylphenol (PVP), PS, polylactic acid (PLA), polydimethylsiloxane (PDMS), polyvinylalcohol (PVA) and benzocyclobutene (BCB) [1,131].

4.2. Substrates for printed electronics

The substrate is a base for the rest of the electronics, additionally, it acts as an electric isolator to separate electric devices from each other. A substrate in printed electronics can be made from synthetic or natural materials; however, different applications and printing methods require different substrate properties, such as flexibility, stiffness, high transparency, surface smoothness, low thermal expansion, heat resistance, low cost, thin and light weight [120]. Traditional substrates for electronics have usually been rigid and heavy, made from materials like silicon or germanium; however, these materials are neither biodegradable, nor recyclable. The development of lighter, flexible and recyclable or biodegradable synthetic polymer substrates has enabled the advancement of new electric applications. Additionally, biodegradable and non-toxic natural materials, such as fibers, resins and proteins, have exhibited insulating properties suitable for substrates [1].

4.2.1. Natural polymeric substrates

Paper substrate is an attractive alternative for printed electronics, due to them being cheap, flexible, environmentally friendly and biodegradable [134]. The disadvantages of paper are its high surface roughness, porosity, vapour permeability and poor moisture resistance. However, the properties of paper can be improved chemically, physically, by coating or laminating [135]. Another suitable natural polymer substrate is nanocellulose, which possesses properties such as high transparency, mechanical strength, heat resistance, low thermal expansion and high surface smoothness [1,20,120].

Other natural biodegradable materials, which can be used as substrates are e.g. silk, shellac, gelatine and starch [20]. Silk is a biodegradable, biocompatible and nontoxic natural protein fiber material, which has been shown to be a promising candidate for use as a substrate. Silk is easy to process and has excellent chemical stability, mechanical properties, and flexibility. Shellac is a natural resin exhibiting biodegradability, high surface smoothness and high solubility in alcohol solvents, which makes it suitable for forming substrate films [1].

4.2.2. Synthetic polymeric substrates

Polymer films are the most popular choice for printed electronic substrates but the manufacturing and operational milieu has to be carefully designed and controlled in order to minimize their surface defects and distortions. PET, polyethylene naphthalate (PEN), PI and polycarbonate (PC) are the most commonly used polymer substrate materials in printed electronics [1]. PET is the most popular polymer substrate, it has high optical transparency, flexibility, solvent resistance, low price and dimensional stability in high temperatures [37]. PEN and PI on the other hand have better heat resistance, but lower transparency and higher cost [120]. PC has high stability, low weight and good mechanical properties, such as rigidity, impact resistance and hardness [136].

PLA, PDMS, PVA, polycaprolactone (PCL), poly-lactic-co-glycolic acid (PLGA), polyurethane (PU), polybutylene succinate (PBS) and polyethylene glycol (PEG) are examples of biodegradable polymers that can be used as substrate materials. PLA is a stiff, transparent polymer, with a slow crystallization rate and low heat resistance, the mechanical properties and heat resistance of which can be improved by e.g. nucleation, change in the stereochemistry or the use of additives [20]. PDMS is a highly elastic biocompatible polymer, which could be used as a substrate in stretchable electronics [1].

In Table 3, the glass transition temperatures, maximum service temperatures and other properties of substrate materials are compared. The maximum service temperature is the highest temperature where the material can be used for an extended period of time without notable problems.

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Table 3. Glass transition temperature, maximum service temperature and other comments about different recyclable and biodegradable substrate materials.

Substrate material	Glass transition temperature (°C)	Maximum service temperature (°C)	Comments
PET	68-80	115-120	Recyclable, excellent water resistance
PEN	118-126	160-180	Recyclable, excellent water resistance, good UV durability, transparent
PI thermoplastic	240-260	221-241	Recyclable, expensive, excellent water resistance, excellent UV durability
PC	142-158	101-116	Recyclable, excellent water resistance, transparent
PLA	52-60	45-55	Recyclable, biodegradable, good UV durability, transparent, high renewable material content
PCL	(-72)-(-59)	40-50	Recyclable, biodegradable
PLGA	44-54	45-55	Recyclable, biodegradable, expensive, good UV durability, transparent
PU thermoplastic	77-107	65-78	Recyclable, biodegradable, excellent water resistance, transparent
Paper	47-67	77-130	Recyclable, biodegradable, high renewable material content
Starch	10-20	60-80	Recyclable, biodegradable, high renewable material content
Silk	77	77-87	Biodegradable, expensive, high renewable material content

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5. Characterization of inks and substrates

5.1. Ink

Ink properties such as viscosity, surface tension, particle size and solid content have large impact on the printed electronics. To elaborate, the viscosity of an ink specifies the resistance against the flow at a specific shear rate. As listed in Table 4, different viscosities are required for different printing processes. The viscosity of an ink can be modified without altering the properties of the ink; however, it is challenging to keep the same electrical properties with changing the viscosity [131]. Increasing the temperature decreases the viscosity of the ink while the solvent evaporation increases the viscosity [137,138]. Flexography, gravure and inkjet printing use low-viscosity, liquid, inks, while offset, screen and pad printing use high-viscosity, paste-like, inks [115].

There is a tension at the surface between a liquid and a gas, because of the asymmetric attractive force between the molecules, this phenomenon is called surface tension. Polar liquids usually have high surface tension, and nonpolar liquids low. An increase in the temperature or increase in solid content can decrease the surface tension of an ink [137,138]. The surface tension of the ink is important for the formation of drops, which also affects the interaction between the ink and the substrate [139].

The functionality of metal particle inks improve with decreasing particle size [115]. Decreasing the particle size increases the surface area and increases the amount of stabilizing agents required. Additionally, smaller particle size in the ink cause a high surface to volume ratio, which leads to the requirement of lower sintering temperatures. Small particle size and uniform size distribution in the ink produce higher viscosity inks and denser printed patterns. Additionally, different printers have different particle size requirements [138].

Higher conductivity can be achieved by using inks with higher solid content. An increase in solid content also leads to a decrease of the viscosity under shear stress, allowing the ink to flow more smoothly from one surface to another and still preventing excessive spreading of the ink after printing. The rheological behaviour of the ink can thus be tailored by changing the solid content of the ink [138]. The ink properties and capabilities of gravure, flexography, offset, screen, inkjet and aerosol printing are compiled in Table 4.

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Table 4. Ink characteristics for different printing methods.

	Gravure	Flexography	Offset	Screen	Inkjet	Aerosol
Viscosity (Pa.s)	0.01-1.1	0.01-2	20-100	0.1-1000	0.001-0.05	0.001-2.5
Surface tension (mN/m)	41-44	28-38	30-37	30-50	25-50	10-20
Layer thickness (μm)	0.1-8	0.04-2.5	0.5-2	0.0015-100	0.05-2.0	0.001-10
Feature size (μm)	70-80	80	10-50	20-100	20-50	5-10
Maximum particle size (nm)	15000	15000	10000	1/10th of mesh opening	1/10th of nozzle diameter	1/10th of nozzle diameter
Maximum preferred particle size (nm)	3000	3000	1000	100	50	50
Maximum solid loading (wt-%)	30	40	90	90	20	55 (ultrasonic automization) 75 (pneumatic automization)

5.2. Substrate

Depending on the application for the printed electronics, the substrate may require properties, such as flexibility, high light transmittance, low surface roughness, light weight, low thermal expansion, stiffness, heat resistance, low cost and low thickness [120]. The print quality is affected by the surface roughness and porosity of the substrate [140]. A high porosity and surface roughness can cause disconnected and nonuniform conductive patterns, due to ink penetration. The disconnection of the conductive particles cause an increase in resistivity and decrease in conductivity. Nevertheless, this can be avoided by using a coating for the substrate, increasing the amount of conductive ink used or printing wider patterns [135,141]. Additionally, the impact surface roughness has on the conductive properties of the pattern are contingent on the properties of the conductive ink [130].

The surface roughness can be analysed by studying the surface topography and the cross-section of a sample. An atomic force microscope (AFM) or scanning electron microscope (SEM) can be used to analyse the surface topography [142]. SEM can additionally be used to study the cross-section of a sample after cutting the sample using a focused ion beam (FIB) instrument. Other topographic measurement devices can also be used to study the surface roughness. The porosity can be examined using a print penetration test or a mercury porosimeter [130].

The performance of electronics is also affected by the surface energy and capacity of absorption of a substrate. The resolution of the printed pattern, ink penetration and ink film thickness are affected by these surface properties. A decrease in porosity, decreases the absorption, and a decrease in capillary absorption, decreases the spreading of the ink laterally, which on the other hand allows the printing of more accurate patterns. Substrate properties, such as these can be improved for printed electronics by using e.g. chemical modifications, physical modifications or coatings [135]. The surface energy can be calculated by testing several known liquids, such as water, ethylene glycol and diiodomethane, on the substrate using an optical tensiometer [130].

The dimensional stability of the substrate affect how the substrate reacts to changes in the environment. Cracks and discontinuity in the printed patterns can be caused by poor dimensional stability. The dimensional stability can be analysed by exposing the printed device to different environmental conditions, such as increasing humidity and temperature and study the changes in the topographic and electrical properties [130]. Furthermore, thermal stability and resistance are often required from a substrate for printed electronics, due to the need for high sintering temperatures for certain inks. The thermal stability can be tested by placing the substrate on a hotplate and analysing the changes in dimensions and colour [142].

Additionally, in some electronic applications it is important to know the dielectric properties of the substrate, the dielectric constant and dissipation factor, in order to achieve the required precise functionality. The dielectric constant of the printed device has to be low in order to prevent degradation in the signal, due to the need for high-speed signaling in electronics. The dielectric properties of a substrate can be examined using methods, such as a microstrip T-resonator, microstrip ring test or parallel plate capacitor [142,143].

5.3. *Ink-substrate interaction*

The quality of the printed pattern can be evaluated by the printability, resolution, shelf-life and adhesion to the substrate [39]. Printed electronics additionally require the ability to form highly conductive patterns, which is achieved by a homogeneous distribution and direct contact between conductive particles in the printed pattern [103, 138]. The print quality is greatly influenced by the interaction between the substrate and the ink during and after the printing process. Ink-substrate interactions include drop impact, spreading, wetting, solvent evaporation rate and penetration, in addition to the drying and ink particle merging on the substrate. The interaction between the ink and the substrate include three stages, it begins with the ink drop impact on the substrate, continues with the wetting and spreading of the ink on the substrate and finally the ink achieves equilibrium with the substrate and environment. The properties of the ink and the substrate, in addition to the environmental conditions, strongly influence this interaction process [137,139].

The print quality is mostly affected by the equilibrium wetting, which can be determined by the contact angle (CA). The CA of a drop on a surface is the angle between the tangents of the liquid-air and liquid-solid interfaces. The surface tension of the liquid, the surface energy of the substrate and the environmental conditions, determine the CA. If the surface energy of the substrate is higher than the surface tension for the ink, the CA is small with an increased spreading of the ink. On the other hand, a high CA with decreased spreading can be achieved by using a substrate with a lower surface energy than the surface tension of the ink. A higher CA with decreased spreading results in thicker patterns with higher resolution and a decrease in the line width [115,137,139].

The contact angle changes when it is observed over a longer time. An ink drop loses mass due to evaporation, resulting in a decreasing CA over time. It is important that the evaporation rate is not too fast and allows for wetting and spreading of the drop. It should not be either too slow with exaggerated spreading or too slow drying. A low advancing CA is required when depositing individual ink drops on a substrate to form a uniform level high quality pattern. Commonly the advancing CA is larger than the receding CA for most combinations of inks and substrates, the difference between these is the CA hysteresis (CAH) [139]. If CAH is high or CA is low, it is probable that the receding CA is zero [137].

Factors affecting CAH are, porosity, surface roughness, chemical heterogeneity and strong attractive interaction between ink and substrate. Surfaces with high porosity absorb ink [115]. The surface roughness can increase the wetting and decrease the CA, if the CA is lower than 90°. On the other hand, if the CA is higher than 90°, the surface roughness restricts the wetting of the surface. CAH is also increased by surface roughness. Additionally, a substrate with a high surface roughness and porosity can cause discontinuity in the printed conductive pattern, which would cause a printed electronic device not to function [103]. The effects of surface roughness can be minimized by surface treatments or substrate patterning. Chemical heterogeneous materials have areas that are more wettable and other areas that are less wettable, the receding CA is usually on the most wettable areas, while the advancing CA is usually on the least wettable area. A drop and a substrate that do not wet, but have a strong attractive interaction, can cause pinning of the edge of the drop [137]. Additionally, high viscosity inks are usually more reluctant to spreading compared to low viscosity inks [139].

5.4. Post-printing methods for device performance

It is often necessary to use some kind of post-print treatment in order to acquire the optimal properties, such as conductivity, for printed electronics. These treatments can be based on physical or chemical reactions and aim to remove solvents and additives from the ink, in addition to improving the morphology and microstructure of the printed pattern [131]. Heat, pressure, plasma, laser, electrical voltage, chemicals, photo- or microwave radiation can be used for post-print treatments. Depending on the ink and the substrate used in the printed electronic device, different kinds of post-print treatments can be used, such as photonic curing, annealing, thermal-, photonic-, microwave-, plasma- or chemical sintering [39]. The sintering process is affected by parameters, such as particle size and shape, temperature, time, radiation energy level and printed pattern thickness. Increased temperature and time, increases the degree of sintering [139].

Thermal sintering is usually accomplished using an oven or hot plate at a specific temperature. It is also to be noted that high temperatures can have a negative effect on the adhesion of ink to substrate and are not suitable for various substrates. Different inks also have varying thermal stability, some inks can endure very high temperatures without a change in conductivity, while others may unexpectedly lose their conductivity at a specific temperature. Thermal sintering is an important post-treatment method for nanoparticle inks. Organic stabilizers are used for metal and oxide particle inks in order to prevent agglomeration; however, after printing, the stabilizers are removed using thermal sintering for the particles to form a continuous pattern. Additionally, by thermally sintering particle-based inks to a temperature below their melting point, the electrical properties can be improved. The melting point and sintering temperature can be decreased by a decrease in the particle size used in the ink, due to an increase in the surface-to-volume ratio [39,139]. MOD inks usually require very high sintering temperatures, which limits the choices for substrates used, and precipitation of metal [132]. Material properties can also be improved by relieving internal stresses in solution-processed semiconductors, by using annealing. Annealing is usually completed using an oven, hot air flow or hot plate. The high temperatures used in annealing, limits its application [131].

The different temperatures required to thermally sinter various nanoparticle inks on glass substrates are compared in Table 5. The silver nanoparticle ink clearly has the lowest resistivity on glass substrate with a relatively low sintering temperature; however, there are other factors affecting the sintering temperature and resistivity as well, e.g. the size of the conductive particle.

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Table 5. Comparison of different sintering temperatures for different nanoparticle inks on glass substrates.

Nanoparticle ink	Sintering temperature (°C)	Resistivity ($\mu\Omega$.cm)
Silver	200	4 [144]
Copper	200	18 [145]
Gold	240	714 [146]
Aluminium	600 (starting from 25 increasing 10 °C/min)	41.2 [147]
Nickel	230	460 [148]
ITO	400	100 [149]

Photonic sintering transfer energy from a light source, such as flash lamps or lasers, to the surface. Fast, high temperature and selective heating can be achieved by photonic sintering. The use of flash lamps enables effective sintering of the printed pattern with high-intensity millisecond light pulses, increasing only the ink temperature without damaging the substrate. Both continuous and pulse lasers can also be used for sintering printed metal nanoparticle patterns. High-resolution patterns can be achieved by using laser sintering, the laser beam can be tuned for different patterns by adjusting the size and intensity [39]. Selective heating methods are practical when using substrates unable to withstand high sintering temperatures. UV-curing on the other hand is commonly used for insulating and chemically stable materials, such as dielectrics or insulators. Without heating, UV-curing can swiftly solidify the ink with sharp edges and smooth morphology [131]. However, plastic absorbs the wavelength range used in UV-curing, this may cause damage to the films [120].

High temperature plasma sintering and low pressure argon plasma sintering are also selective sintering techniques [131]. Low pressure argon plasma is the most commonly used plasma sintering method for printed patterns. The sintering starts from the surface and continues into the bulk, an increase in treatment time, decreases the resistivity. Plasma sintering can be used for electronics printed on plastic substrates due to the ability to use low temperatures but the penetration depth of the plasma limits the possible achievable thickness [39,120].

Microwave sintering is a very fast method for sintering metals. The penetration depth of the radiation is, however, very small (approximately 1-2 µm at 2.54 GHz), which limits the thickness of the printed pattern when using microwave sintering [120]. Metals with high thermal conductivity can still use thicker patterns, due to the thermal conductance, and still form uniform patterns [39,131].

Electrical sintering heats the printed metal pattern using electric current, caused by an application of voltage over the pattern. The printed pattern is required to be somewhat conductive already before the electrical sintering. This sintering method is low temperature and very fast [39].

Chemical sintering is performed using chemical agents in order to provoke nanoparticles to merge at room temperature. Oppositely charged polyelectrolytes are applied to the metal nanoparticles to stimulate a spontaneous process where the nanoparticles merge and create a conductive pattern. The polyelectrolyte can be added onto the substrate prior to printing or onto the printed pattern after printing. Chemical sintering enables the use of substrates sensitive to heat, such as plastics and paper. Negatively charged silver nanoparticles can be chemically sintered using poly(diallyldimethylammonium chloride) (PDAC), as a positively charged polyelectrolyte [150]. Other examples of chemical agents are ascorbic acid, hydrochloric acid (HCl), sodium chloride (NaCl), magnesium chloride ($MgCl_2$) and chloride ions (Cl^-) [39,103,131].

The differences in the resistivity of silver nanoparticle inks on glass and PI substrates using different sintering methods are compiled in Table 6. In addition to the sintering method, substrate and ink, the resistivity depends on the silver particle size, other materials used in the ink and the printed pattern.

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Table 6. Comparison of different sintering methods for nanosilver ink on glass and PI substrates.

Sintering method	Resistivity ($\mu\Omega$.cm) Glass substrate	Resistivity ($\mu\Omega$.cm) PI substrate
Thermal	4.00 [144]	3.60 [151]
Flash lamp	5.59 [152]	3.30 [151]
IR lamp	3.00 [153]	65.50 [154]
Laser	3.41 [155]	4.60 [151]
UV	48.00 [156]	6.50 [157]
Argon plasma	8.73 [158]	15.00 [151]
Microwave	-	30.00 [159]
Electrical	2.47 [160]	17 [161]

5.5. Life-cycle assessment

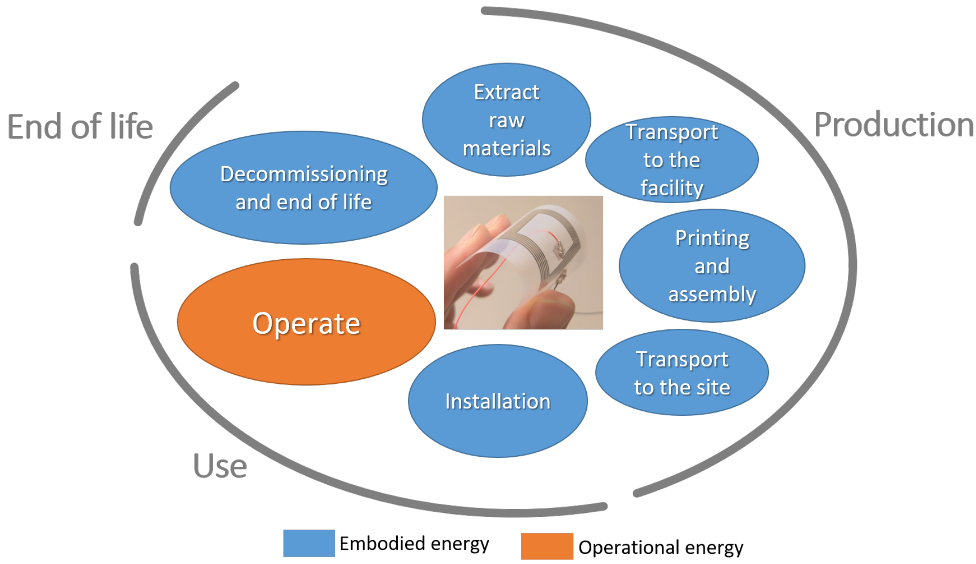

Figure 12. Holistic overview for life-cycle of a printed electronics device. Operational energy is the energy required to use the device while embodied energy refers to the consumed energy by the processes associated with the production and end of life.

In order to address the environmental impacts of the printed electronics, comprehensive assessment throughout the device life, known as life cycle assessment (LCA), is a must. As holistically schematized in Figure 12, LCA of printed electronics provides relevant quantification of the environmental aspects at different stages, i.e. production, use and end-of-life, which are based on the ISO 14040:2006 (Environmental Management-Life Cycle Assessment-Principles and Framework), ISO 14044:2006 (Environmental Management-Life Cycle Assessment-Requirements and Guidelines) and ISO 14067:2018 (Greenhouse gases — Carbon footprint of products — Requirements and guidelines for quantification) [162–164]. These standards complement each other describing the limitations and applications of LCA, data collection and interpretation for the environmental impact assessment [20]. In alignment with these standards, several studies have been conducted which fully or partially covers the life cycle stages. For instance, aging, degradation, environmental and safety issues of polymer films have been studied and explained in detail by Izdebska [115] and Radermacher [165]. Environmental assessments and toxic emission evaluations for printed antennas have been studied by Kanth et al.[166]. LCA and recycling options for photovoltaics were proposed by Espinosa et al. [167], Gong et al. [168], Sondergaard et al. [169] while energy demand and reusability of biodegradable and recycled printed LED foils were investigated by Valimaki et al. [20]. In addition, energy demands, performance and life-time comparisons of OFETs were provided by Vladu [170]. Recent studies by Dinh et al. [171], Yan et al. [172] and Zhang et al. [173] demonstrated the effective printing of carbon based inks on thermoplastic substrates, which are highly recyclable. The leading work of Williams et al. was also a paramount of printable and recyclable sensors and TFTs composed of fully carbon based inks and lignocellulosic substrates [66]. Although there exist limitations, the scientific breakthroughs illustrate the capabilities of printed electronics as recyclable devices with reduced carbon footprints.

6. Conclusions

The present article provides a thorough revision comprised of the manufacturing, material, application and environmental aspects, advantages and limitations of recyclable printed electronics. Based on the trends in industry, science and consumer markets, commonly used substrates, inks, and contact and non-contact manufacturing methodologies are outlined in line with their applications. Despite the limitations in their performance, durability and reliability in comparison with their conventional counterparts, printed electronics offer rapid and affordable manufacturing, and less environmental hazards. Therefore, printed electronics is foreseen to be more accessible and available in our daily lives, which will be one of the gateways towards a digital 21st century. By means of the scientific breakthroughs and urgent need of smart device technologies for the industrial and IoT revolutions, gadgets and devices using printed components are expected to be progressively used not only in consumer electronics but also in energy harvesting and storage, biomedical applications, transportation and constructions, structural and agricultural sensing and monitoring to name a few. Hence, automation, communication of devices, data collection, screening and transfer will be affordably and autonomously carried out with minimum demand for labor, fossil-based energy and bulky electronic systems.

Author Contributions

All authors contributed to the manuscript while J.W. and A.K. conceptualized and coordinated the manuscript writing and editing processes. All the authors have agreed to the present version of the manuscript.

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Data Availability Statement

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Conflicts of interest

The authors declare no conflict of interest.

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