IOP Publishing Flex. Print. Electron. 7 (2022) 043001 https://doi.org/10.1088/2058-8585/ac91de

Flexible and Printed Electronics

OPEN ACCESS

RECEIVED
26 November 2021

REVISED
13 June 2022

ACCEPTED FOR PUBLICATION
14 September 2022

PUBLISHED
5 October 2022

TOPICAL REVIEW

The role of printed electronics and related technologies in the development of smart connected products

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Keywords: flexible and printed electronics, smart connected products, internet of things, additive electronics, Society 5.0

Abstract

The emergence of novel materials with flexible and stretchable characteristics, and the use of new processing technologies, have allowed for the development of new connected devices and applications. Using printed electronics, traditional electronic elements are being combined with flexible components and allowing for the development of new smart connected products. As a result, devices that are capable of sensing, actuating, and communicating remotely while being low-cost, lightweight, conformable, and easily customizable are already being developed. Combined with the expansion of the Internet of Things, artificial intelligence, and encryption algorithms, the overall attractiveness of these technologies has prompted new applications to appear in almost every sector. The exponential technological development is currently allowing for the 'smartification' of cities, manufacturing, healthcare, agriculture, logistics, among others. In this review article, the steps towards this transition are approached, starting from the conceptualization of smart connected products and their main markets. The manufacturing technologies are then presented, with focus on printing-based ones, compatible with organic materials. Finally, each one of the printable components is presented and some applications are discussed.

1. Introduction

Historically, humanity has experienced several critical paradigm shifts that dictate the way society behaves, solves problems, creates value, and develops itself as a whole. Starting from Society 1.0, the hunter-gatherer society, the world has seen the upsurge of agriculture (Society 2.0), industry (Society 3.0), the expansion of information and automation technologies (Society 4.0), and is currently heading toward the establishment of a 'Super Smart Society' (Society 5.0) [1]. As seen in figure 1, five industrial revolutions have been responsible for fueling the developments attributed to modern society, which started in the 18th century with the use of water, steam and fossil fuels to generate mechanical energy [2]. This period became known as the 1st Industrial Revolution (Industry 1.0). In the 2nd Industrial Revolution, electrical energy took over the way mass production factory lines were powered. This was followed by the 3rd Industrial Revolution, which took place in the second half of the 20th century, and was defined by the emergence of automated systems, as well as other advanced digital developments, that brought us one step closer to the world we know today. From this moment onwards, electronics and information technology have been used together to create complex systems and active devices [3]. The economy became based upon computer and internet-aided systems, factories became automated, telecommunications became widespread, and biotechnology emerged [3]. The transition to the 4th Industrial revolution started around 2011 and is still ongoing [4]. This recent technological era is characterized by a significant increase in the interconnectivity of systems, the decentralization of decisions made by such systems (which are becoming ubiquitously distributed and mainly autonomous), and the on-demand availability of computer system resources.

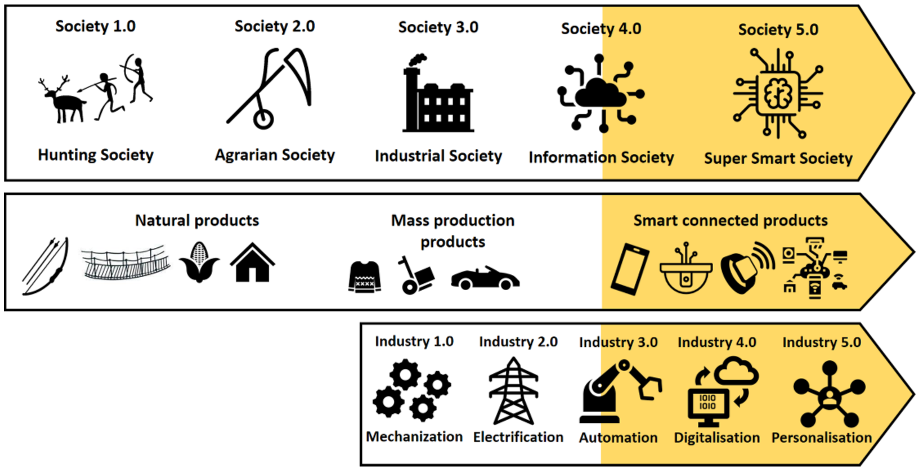


Figure 1. Evolution of Society paradigms and Industrial Revolutions throughout the ages.

In the Industry 4.0, the human is removed from the factory ground to remote areas where control and monitoring tasks are safely performed.

Nevertheless, due to the exponential speed at which technology is currently developing, a novel industrial revolution, Industry 5.0, is already on the horizon and is mainly characterized by the personalization of industry [2]. As a result, in the near future, the human is expected to return to the factory ground and cooperate closely with robots and semi-automated systems. This should occur gradually, as soon as technological advancements, artificial intelligence (AI) algorithms, and safety norms become sophisticated enough to allow for a fluid and safe cooperation between humans and machines. Moreover, Industry 5.0 and Society 5.0 are both aligned by common goals regarding natural environment protection, sustainable development, and social responsibility [5]. This reflects in the design, materials, manufacturing technologies, and life cycle of the products developed by the Industry 5.0 and directed at the Society 5.0.

There is a close relationship between technology, industry, and society where technological developments continuously feed the growth of the remaining sectors. With the digitalization and personalization of society and industry, smart connected products (SCPs) have taken over the markets worldwide [6]. These products are characterized as physical devices that encompass smart components and connectivity elements [6, 7]. The upsurge of novel SCP, much thanks to research and development (R&D) efforts and industrial developments, ends up defining the way products and services reach the markets and continuously sculpt our society [7].

1.1. The dimensions of SCPs

Due to their high demand, SCP are flooding the current markets. They can be classified as consumer products that comprise some sort of intelligence-generating technology, and for this purpose, they include sensors and/or actuators (that gather data or use data to generate a response), electronic components and/or advanced processing units (for data processing and analysis), and they can also include interfaces or communication units (to exchange the gathered information) [6]. They also need to be powered, and in some cases can have memory units to temporarily store data before it is communicated.

Thus, as evidenced in figure 2, SCP rely on the integration of printable sensors and actuators, active touch and gesture sensors, printable processing circuits, radio-frequency identification (RFID) antennas, near-field communication (NFC) labels, organic photovoltaic (OPV) chargers, organic thermoelectric generators (OTEGs), and other green energy generators, printed memories and batteries, and organic displays (amongst other components). These components will become the pivotal physical building blocks of this transition.

Given the sustainability goals for the future, some parts of SCP are being manufactured using new materials and manufacturing technologies, capable of responding to the market needs in terms of functionality, simplicity, sustainability, and affordability. As a result, research in additive electronics technologies such as printed electronics (PE), 3D PE, in-mold electronics (IME), 3D-molded interconnected device (MID) devices, and organic large-area electronics (OLAE) is gaining traction and holds promise to revolutionize the future of electronics applied to SCP [8, 9]. PE and OLAE are emerging technologies fueled by research on a new class of materials that can be used in large-area and high-volume deposition or printing/patterning of electronic components and devices [9–11]. The use of additive electronic manufacturing strategies allows for rapid development of case studies, and scale-up of new devices, while economizing materials [12–14]. Some physical components such as sensors, actuators, antennas,

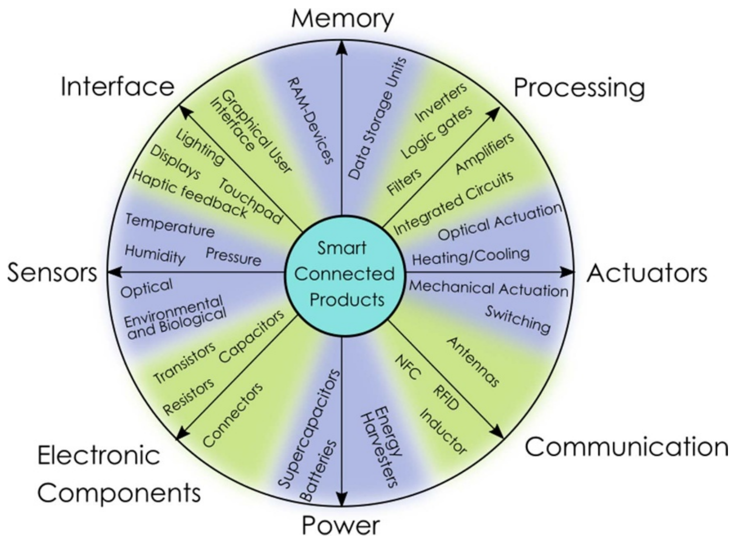


Figure 2. Diagram depiction of elements integrated into the production of SCP. Highlighted in green are the elements already achievable through PE technologies and approached in this review.

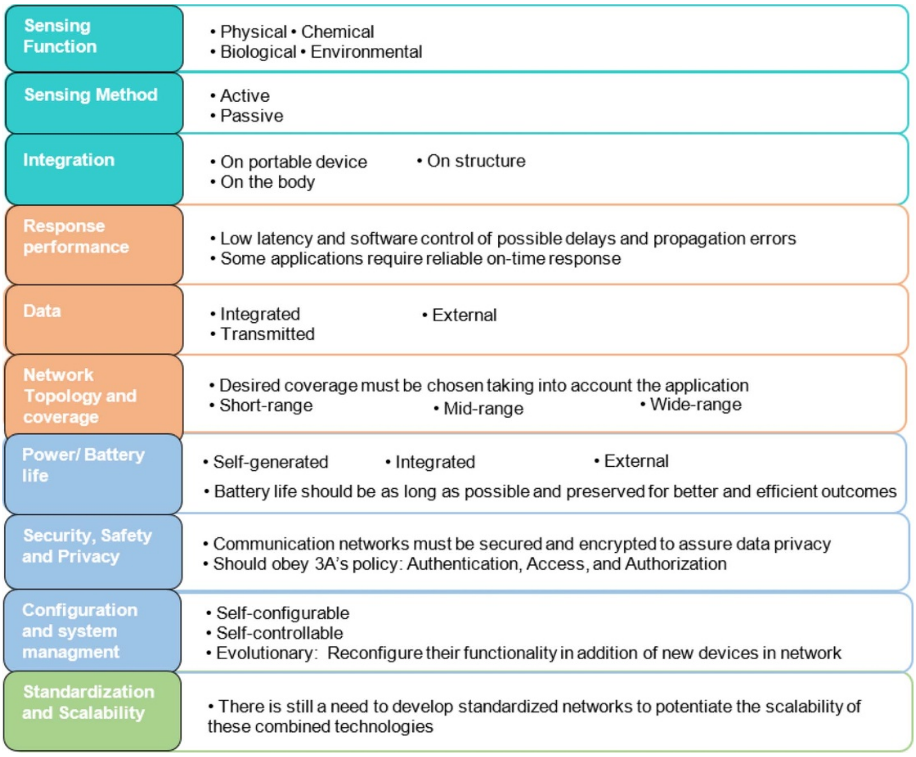
displays, energy harvesters, and organic batteries can already be obtained through PE and OLAE, however many of them still present performance limitations [9]. For instance, printed high-performance transistors obtained from roll-to-roll (R2R) industrial processes still suffer from a lot of variability and low electronic mobility value [15, 16]. For this reason, PE commercial applications usually adopt hybrid systems, referred to as flexible hybrid electronics (FHE), i.e. devices obtained by the integration of conventional (rigid surface mount devices (SMDs)) and PE [11]. This technology integrates PE with traditional or thinned silicon (SI) IC chips and other components to produce affordable, mechanically flexible, and relatively complex and robust circuit systems [11, 15]. By merging different manufacturing technologies, device architecture designs, and integrated systems, new functionalities, applications, and new markets are set to emerge and establish themselves. Throughout this review, we will refer to PE as an encompassing term that includes OLAE.

The design of SCP usually develops around the main sensing function, which comprises specific sensors and/or actuators. The main factors to be taken into account during the development of novel SCP are schematized in figure 3 [17].

1.2. SCPs for Society 5.0

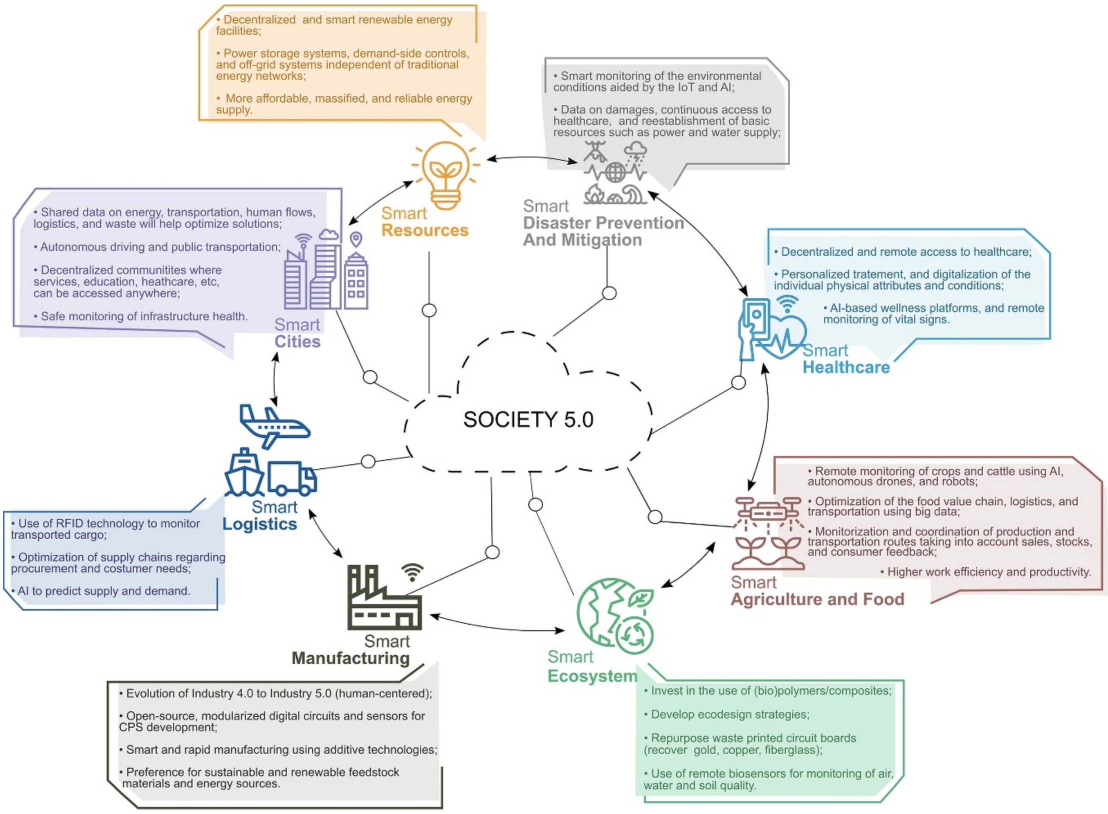
Along with the exponential increase of computing power and communications technologies, and the near-omnipresence of smart products around humans, people themselves are expected to become the central element in this so-called ongoing digital transformation. Hence, connections between ‘people and things’ are rapidly leading to the establishment of a new societal archetype, known as Society 5.0 [18]. Through the auscultation of the needs of the society of the future, some predictions can start to be formulated, and R&D efforts in innovative fields accelerated.

Figure 4 shows the main fields involved in the Society 5.0 paradigm and how the SCP will empower each one of these sectors [19]. SCP are expected to help connect the different services, including the decentralization of education and healthcare access, which are sometimes out of reach of the communities located further from the big city centers. These systems will also help monitor the infrastructure health of buildings and optimize transportation, logistics, and waste management. Disaster prevention, mitigation, and first-aid response will also benefit from SCP implementation for monitoring and data gathering. The exploitation of resources, especially renewable ones, is also expected to become more efficient, decentralized, massified, and reliable. Regarding the ongoing revolution of Industrial manufacturing, in the future, the role of the human operator will become more centralized and the machinery will be aware of the human presence and able to cooperate with it. The investment in additive technologies is prone to increase continuously, and renewable feedstock materials and energy sources will be preferred. Agriculture and food production are also sectors that will benefit exceptionally from the implementation of these connected technologies. Taking advantage of smart logistics and the ubiquitous use of intelligent and reprogrammable RFID tags, the consumables value chain will be enriched, and the circular economy model will be applied, whenever possible [5]. Their large-scale integration with the Internet of Things (IoT), AI algorithms, and blockchain technologies will allow the interconnection of services, markets, and institutions, as seen in figure 4 [20].



Sensing Function	• Physical • Chemical • Biological • Environmental
Sensing Method	• Active • Passive
Integration	• On portable device • On structure • On the body
Response performance	• Low latency and software control of possible delays and propagation errors • Some applications require reliable on-time response
Data	• Integrated • External • Transmitted
Network Topology and coverage	• Desired coverage must be chosen taking into account the application • Short-range • Mid-range • Wide-range
Power/ Battery life	• Self-generated • Integrated • External • Battery life should be as long as possible and preserved for better and efficient outcomes
Security, Safety and Privacy	• Communication networks must be secured and encrypted to assure data privacy • Should obey 3A's policy: Authentication, Access, and Authorization
Configuration and system managment	• Self-configurable • Self-controllable • Evolutionary: Reconfigure their functionality in addition of new devices in network
Standardization and Scalability	• There is still a need to develop standardized networks to potentiate the scalability of these combined technologies

Figure 3. Compartmentalization of the dimensions associated with smart connected products.



- Decentralized and smart renewable energy facilities;
- Power storage systems, demand-side controls, and off-grid systems independent of traditional energy networks;
- More affordable, massified, and reliable energy supply.

- Smart monitoring of the environmental conditions aided by the IoT and AI;
- Data on damages, continuous access to healthcare, and reestablishment of basic resources such as power and water supply;

- Decentralized and remote access to healthcare;
- Personalized tratement, and digitalization of the individual physical attributes and conditions;
- AI-based wellness platforms, and remote monitoring of vital signs.

- Remote monitoring of crops and cattle using AI, autonomous drones, and robots;
- Optimization of the food value chain, logistics, and transportation using big data;
- Monitorization and coordination of production and transportation routes taking into account sales, stocks, and consumer feedback;
- Higher work efficiency and productivity.

- Invest in the use of (bio)polymers/composites;
- Develop ecodesign strategies;
- Repurpose waste printed circuit boards (recover gold, copper, fiberglass);
- Use of remote biosensors for monitoring of air, water and soil quality.

- Evolution of Industry 4.0 to Industry 5.0 (human-centered);
- Open-source, modularized digital circuits and sensors for CPS development;
- Smart and rapid manufacturing using additive technologies;
- Preference for sustainable and renewable feedstock materials and energy sources.

- Use of RFID technology to monitor transported cargo;
- Optimization of supply chains regarding procurement and costumer needs;
- AI to predict supply and demand.

- Shared data on energy, transportation, human flows, logistics, and waste will help optimize solutions;
- Autonomous driving and public transportation;
- Decentralized communitites where services, education, heathcare, etc, can be accessed anywhere;
- Safe monitoring of infrastructure health.

Figure 4. Fields involved in the Society 5.0 paradigm and description of how the SCP will empower each one of the sectors.

As a result, the manufacturing pathways of each one of these physical components is approached in his review article, which is organized as follows. Section 2 critically discusses the printing-based technologies that can be used to obtain SCP. PE and other integrative solutions for stretchable electronics are presented. In section 3, the printability of each one of the SCP building blocks is evaluated and each one of them is presented along with some literature examples. Section 4 is the concluding section, where the benefits and challenges of these technologies are briefly discussed along with some future perspectives.

2. Technologies for SCP

In the following section, manufacturing and integration techniques for SCP will be discussed, as presented in table 1.

When there is no need for the electronics to be developed over a rigid substrate and they are instead created directly over the final structure of the device, we are in the presence of PCB-less electronics, which presents several advantages in terms of design freedom, material economization, and allows for thinner and lightweight products to be obtained [21]. Thanks to these technologies, a new generation of PCB-less electronic applications is emerging, as seen in figure 5.

2.1. PE

Although PE has recently been gaining a lot of interest in the field of product development it is not a novel technology, and its origin dates back to the 1950s [28]. Back then, gravure and screen printing techniques, both additive manufacturing methods, were already deemed by researchers as promising technologies for the production of conductive wiring [28]. Even though subtractive techniques, characterized by the use of chemical etching methods and several other steps, have been the ultimate methods used to produce printed circuit boards (PCBs), PE has become an alternative to traditional PCB technologies, relying on functional printable inks and pastes that can be obtained from organic and inorganic materials [29]. These inks are not only easier and more economic to formulate, but also exhibit intrinsic stretching ability. Moreover, the technologies used to deposit them onto substrates are mostly additive processes that save up material [30]. With the widespread development of connected applications and the increasing need for the development of seamless and conformable sensing and actuating systems, innovative materials and techniques are being preferred. These techniques should allow for rapid prototyping, large-scale production, multiple layers deposition, deposition over non-planar surfaces, reproducibility, and good overall performance of the resulting devices [30].

Printing technologies for the development of electronic devices can be divided into contact and non-contact printing [30]. Contact-printing technologies usually require stamps, screens, molds, or masks tailored specifically to define the printing patterns, and include screen-printing [31], gravure-printing [32], flexographic-printing [33], offset printing, and soft-lithography techniques, among others. Non-contact printing techniques do not demand accessory equipment, nor waste as much material as the abovementioned techniques. Besides, the printing patterns are usually defined through software and can be edited without constraints, apart from the printing equipment resolution, thus allowing for large-scale production of 'mass customized' products. Some non-contact printing technologies are inkjet printing (IJP), electrohydrodynamic jet printing, and aerosol printing. These non-contact printing technologies, along with soft-lithography methods, and some 3D printing techniques can be used to directly print electronics over curved and three-dimensional surfaces [34].

Another emerging approach in the manufacture of PE is the automated printing of devices resorting to 3D printing technologies (using e.g. extrusion, binder jetting, photopolymerization, powder bed fusion, direct energy deposition, and sheet lamination processes) [35, 36]. This involves one-step multi-material printing that will soon be possible through the symbiotic combination of diverse 3D printing technologies and powerful software development. This set of cooperating technologies is called hybrid 3D printing and vows to embed the 3D printing of solid conductive wiring into the device structure, which will be simultaneously printed, allowing for the creation of 3D meanders and interconnects. As a result, fully functional and interactive sensor and actuator devices can be obtained in a one-step procedure [35, 37]. Table 2 summarizes the main printing technologies and their main characteristics.

2.2. FHE

FHE demands the use of complementary technologies for printing and integrates both traditional electronics and PE. It is backed by the premise of 'print everything you can, and place what you can't' [57]. The gradual replacement of rigid materials with flexible, inexpensive, organic, hybrid, and composite ones, has potentiated the design of several components through additive electronics techniques. Nonetheless, the higher performance of traditional electronics has not yet been met, and, in many cases manufacturers integrate SMDs directly over the flexible substrates. SMD encompass active, passive, and electromechanical or electrical components that have been used in microelectronics for decades. They are already inherently small and thin and are adequate to be integrated over flexible and stretchable substrates as long as the right integrating approaches are used. Nonetheless, due to their bulky nature, conventional IC chips cannot be integrated into flexible circuits [58]. Hence, when seamless integration of IC

Table 1. Additive electronics technologies, including materials and manufacturing pathways.

	Electronics production				
	Traditional Electronics	Printed electronics (PE)	Flexible Hybrid Electronics (FHE)	Stretchable and Conformable Electronics (SCE)	Structural Electronics
Materials	Silicon, Ceramics, Glass, Solder, Copper, Epoxy (Flame Retardant, FR-4)	Functional inks, low-temperature organic polymers (inks, pastes), and flexible substrates	High and low-temperature processing materials. Rigid parts integrated with soft flexible counterparts	Use of flexible and stretchable inks or other conductive materials integrated with stretchable and conformable substrates	High and low-temperature processing materials. Flexible and stretchable substrates integrated with rigid structural parts (e.g. injectable plastics)
Manufacturing Techniques	Photolithography, etching, ablation, high vacuum deposition, screen printing, etc.	Printing on a polymer, textile, paper, cork, skin, etc. Low-temperature processes	Printing techniques combined with pick and place and encapsulation techniques	Printing techniques, stretchable polymer-matrix composites with reinforcement of interconnections, and meander-like designs	3D Molded Interconnected Devices (3D-MID), in-mold electronics (IME), 3D printed electronics

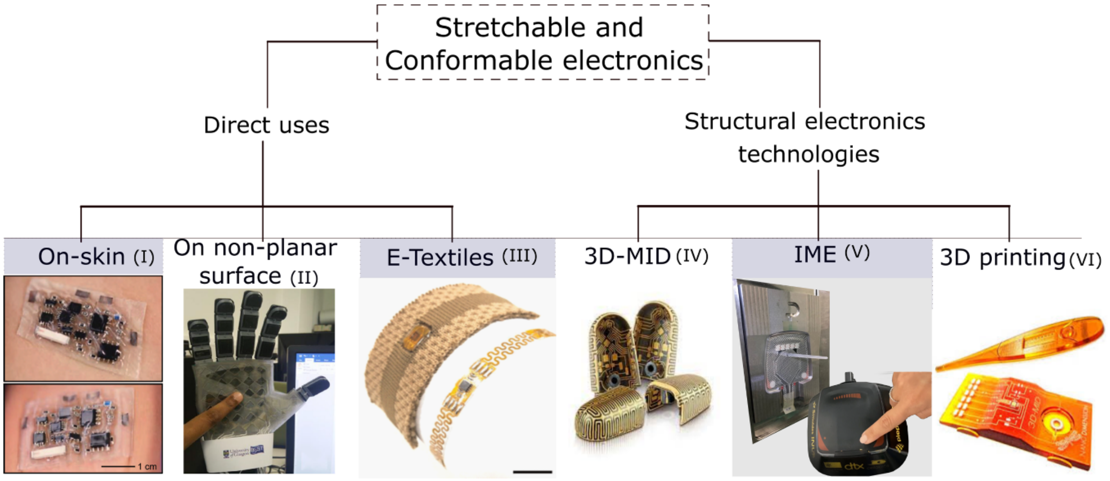


Figure 5. Device integration. (I) On-skin electronics (e-skin device). From [22]. Reprinted with permission from AAAS.. (II) Nanowire FET pressure sensing array on a robotic hand. Reproduced from [23]. CC BY 4.0. Flexible and stretchable electronics embedded on textile for vital signs monitoring (e-textile). Reproduced from [24]. CC BY 4.0. Laser direct structuring of 3D-Molded interconnect devices (3D-MID). Reproduced with permission from [25]. © Copyright Distributed Micro Technology Ltd 2022; (V) In-mold electronics (IME) demonstrator developed in the framework of the Molded Electronics project by DTx, Celoplás, Bosch, PIEP, Minho University, Simoldes and INL consortium. Reproduced with permission from [26]. (VI) 3D printed embedded electronic devices created by Nanodimension® LDM^{TM} technology. Reproduced with permission from [27].

chips and mechanical flexibility of the non-printable components are demanded, IC thinning is required [59]. This process, also known as die thinning (or simply dicing), involves the dicing of the chip channels after the wafer has been attached to a dicing tape for supporting the chips. Thicknesses below 50 μm are achievable, deeming the thinned IC chips (also known as ultra-thin chips) compatible with flexible substrates, as represented in figure 6 [11, 59, 60].

To precisely align the SMD and ultra-thin chips in position, a pick-and-place (PnP) tool is usually required. In many cases, for large-scale industrial production, the integration of rigid components by PnP is conducted in a parallel production line to R2R printing, sometimes leading to damage or contamination of the substrates. Moreover, this demands that the production facilities are equipped with both R2R and PnP technologies, which increases production costs [11, 59]. As a result, R2R-capable PnP machines have emerged and are already commercially available [61]. To help with the accurate alignment of components over conductively wired foils,

Table 2. Printing characteristics of the previously described technologies. [9] John Wiley & Sons. © 2021 Wiley-VCH GmbH.

		Line width resolution (μm)	Ink/Paste viscosity (cP)	Ink waste	Mask	References
Screen printing		30–100	500–5000	High	Yes	[38]
Flexography		20–80	10–500	High	No	[38]
Gravure printing		2–200	50–200	High	No	[32, 39, 40]
Reverse gravure printing		5–200	10–200	High	No	[40–42]
Gravure-offset printing		20–70	10–50	High	No	[40, 43]
Reverse offset printing		0.5–1	1–10	High	No	[44–46]
Soft-lithography	μCP	0.1	—	Low	Yes	[47]
	NI	0.01	—	Low	Yes	[48]
Transfer printing		~3	50–500	Low	No	[49]
Inkjet printing		20–100	1–30	Low	No	[28]
EHD jet printing		0.2–1	1–1000	Low	No	[50, 51]
3D printing	FDM	10–200	$>10^5$	Low	No	[36, 52, 53]
	DIW	1–100	10^4–10^5	Low	No	[54]
Aerosol jet printing		10–20	0.5–2000	Medium	No	[55, 56]

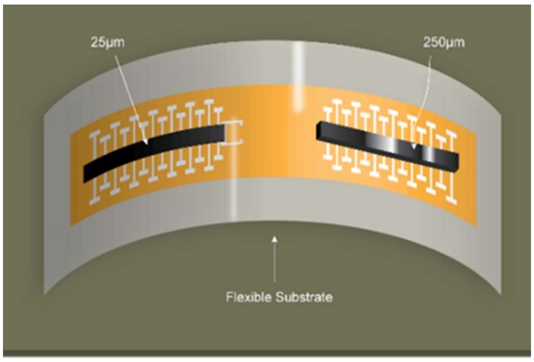


Figure 6. Illustration of a flexible substrate comparing the mechanical flexibility of a 25 μm thinned silicon die IC chip with a 250 μm conventional one (not in scale).

some systems use image recognition tools that assure the perfect positioning between parts and substrate [61]. Since thinned chips are extremely fragile, their mounting process might demand extra concerns, thus, they are usually embedded in packaging film or foil (system in a foil, SiF), which protects them from being damaged during manipulation [59, 60, 62].

To attach the ultra-thin chips and other rigid elements, the available methods are soldering, conductive adhesives, or printable inks [60]. Generally, the use of solder (tin-lead) for flexible electronics is restricted since it demands high temperatures (>250 °C) [63, 64]. As a result, some solder alternatives that present lower reflow temperatures (<175 °C) based on indium, bismuth, and silver have emerged [65]. Low melting temperature solders (<150 °C) have also been developed and are usually based on tin-indium (Sn-In), tin-bismuth (Sn-Bi), and Bi-Sn-In ternary alloys [66–69]. In some cases, their electrical properties can be further improved by reinforcing them with nanofillers such as multi-walled carbon nanotubes (MWCNT), single-walled carbon nanotubes (SWCNT) [70], and graphene [71]. Conductive adhesives are another alternative and, depending on the nature of their conductivity, they can be classified as isotropic (conduct in all directions) or anisotropic (only conduct in the z-plane direction). They are composed of conductive fillers and small particles or flakes of polymer binders that can be transferred through screen-printing or using a stencil. American Semiconductor, Inc. was the first manufacturing facility to install an R2R manufacturing line capable of using the Semiconductor-on-Polymer™ (SoP) technology to directly integrate thinned IC chips in a process called SoP chip scale packaging (CSP) [72]. The SoP CSP technology is currently the main enabler of the FHE developments, granting the maximum flexibility of electronics through the attachment of ultra-thinned IC chips into the smallest packages possible. Moreover, this technology allows the simultaneous integration of both flexible conductive interconnects and other discrete thinned semiconductive components.

An alternative technology to manufacture flexible IC chips is based on thermal evaporation of thin metal oxide films and is already commercialized by PragamatIC® [73]. This is an ultra-low-cost procedure with very high throughput and repeatability. Although the applications are still stuck to low-power ones, such as smart packaging and labeling [74], this is a very promising alternative for the flexible SoP market and, consequently to the growth of the FHE spectrum of applications. Another recently developed packaging method for FHE is based upon a novel fan-out wafer-level packaging methodology and has been described by Fukushima *et al* [75]. This method differs from conventional ultra-thin die IC-chip integration techniques because the Si dielets are heterogeneously embedded in Poly(dimethylsiloxane) (PDMS), and interconnected through high-density wirings, formed at wafer-level processing. This renders them an even higher bending curvature radius and higher durability [75].

With the support of FHE, it is possible to develop large-area, low-cost, and high-performance

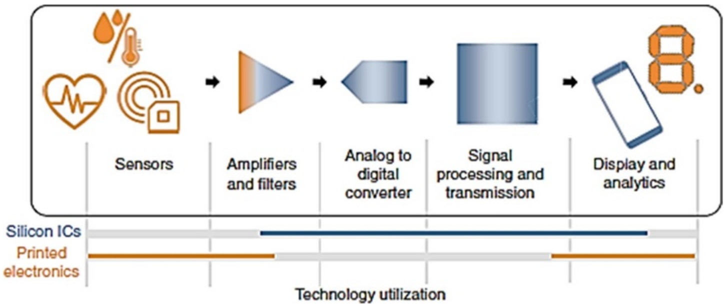


Figure 7. Compromise between printed and traditional electronics in implemented FHE applications. [15] John Wiley & Sons. © 2019 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

flexible devices for a multitude of applications. Printed sensors and displays are already a well-established reality and highly compatible with large area applications [76–80]. Semiconductor processing, however, is still under development and large-area scaling is yet to be optimized. Figure 7 brings insight as to which components can be fully printed, which ones cannot, and which ones can utilize both printing and traditional technologies.

As described above, the use of ultra-thinned chips, accompanied by the growth of the SiF and SoP markets, and other alternatives, are therefore demonstrated technologies that prove the feasibility of integration of Si in FHE. Nonetheless, to integrate these components in printed circuits, they must be adequately interconnected with the remaining substrates and materials. This step can be challenging and can be tackled by making the right design, architecture and material choices for the interconnects, as approached in the following subsection [81].

2.3. Integrative solutions for stretchable electronics

Even though PE, and FHE technologies are being implemented together to achieve new devices, the combination of materials with different flexural moduli and stretching ability often leads to impaired performance and low deformation repeatability of the systems. Consequently, some solutions have been engineered to better integrate the inherently rigid parts, allowing different materials to be efficiently assembled on top of non-flat surfaces or into 3D structures, without loss of functionality. Thanks to their shape conformability and the elevated number of degrees of freedom of their structure, the resulting electronic devices are often called stretchable and conformable electronics (SCE) and are particularly important for areas such as bionics, prosthetics, soft-robotics, e-skin development, and wearables [36, 81–87].

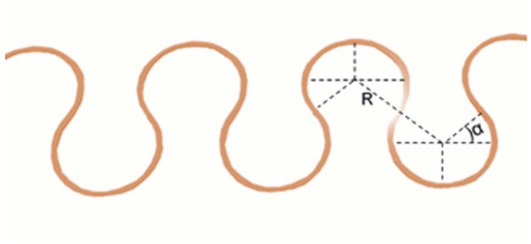


Figure 8. Parameters of the applied circular-based horseshoe pattern. R is the radius of the horseshoe and α is the rotational angle.

To promote the development of SCE and integrate intrinsically stiff components (such as IC chips, and other SMD), some design strategies can be adopted. Hence, patterned printed interconnects can be used to make sure the conductivity is maintained throughout the use of the device. To allow for the stretching and conforming ability, the design of the electrical circuit interconnects, especially in the transition areas between rigid components, should be projected in patterns that can be zigzag or meander-like. These interconnects are projected in the same plane as the electronic SMD and stretchable inks are normally preferred to print the circuitry [58, 88]. One of the most popular and efficient patterns is commonly known as the horseshoe shape [89]. This pattern was first implemented in 2006 by Brosteaux *et al* who designed stretchable interconnects with a radial structure (R) that resembled a horseshoe (figure 8) [90].

This work was a pioneer in successfully implementing this geometry onto the circuitry of an MID. The results proved this design allowed for enhanced stretchability, with maximum elongation of 100% of the original shape. It was also proven that the elongation ability increased with the horseshoe radius. Later, Brand *et al* also studied the reliability of integrating rigid islands using copper meander interconnects printed on PDMS and supported/ encapsulated by a combination of different polymer foils (Polyethylene terephthalate, PET/ Polyethylene naphthalate, Polycarbonate, PC, and Polyimide, PI). When compared with conventional patterns, the meander-like interconnects prevented stress accumulation and material fatigue in key transition areas and assured the maintenance of the functionality of the devices during many stretching cycles [91]. Moreover, while non-encapsulated meanders

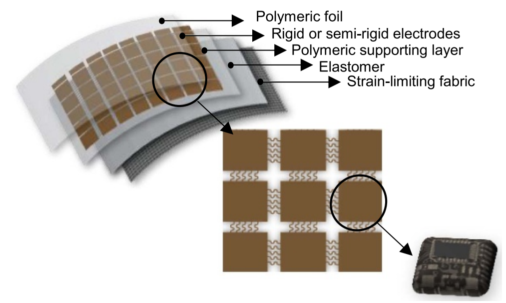


Figure 9. Stretchable and conformable modular circuit concept. The encapsulating layers that grant the circuit higher stress and strain resistance are also depicted.

only sustained around 100 cycles of 10% elongation deformation, the ones encapsulated with PI successfully underwent 120.000 cycles (of 10% elongation) with only 1% of relative loss of efficiency [91]. An example of a modular and encapsulated stretchable and conformable array is depicted in figure 9.

Another way of providing the conforming ability to substrates, interconnects, and electrical circuitry, is to integrate SMD in a dispersed way and print the interconnects on top of pre-strained substrates [58]. This method is efficient for wearables and electronic skins, since when the strain applied to the substrate is released, an out-of-plane wrinkled structure capable of stretching that resembles our knuckles, is obtained [82]. The technique, also known as 'buckling', has been used almost since the emergence of the PE field, as we know it, and is praised for its simplicity and compatibility with a wide variety of materials (both organic and inorganic, printed and traditional), and printing and deposition technologies [92, 93].

Independently from the selected design strategy, the materials used to develop circuitry and connectors can be divided into two fundamental approaches, which are described below [94].

2.3.1. Use of conductive flexible composites

By dispersing conductive fillers, such as electrically conductive particles, flakes, and/or wires, concentrated at their percolation threshold in the elastomeric matrixes, conductive pathsways are efficiently created [94]. Frequently used filler materials include carbon black (CB) particles [95], carbon nanotubes (CNTs) [96], silver nanoparticles (AgNPs) and nanowires (AgNWs) [97, 98], and other metallic particles [99].

As an example, Yellapantula *et al* created flexible sensor arrays for robotic applications using Ecoflex and PDMS as matrix and a composite made from CB particles, PDMS, and Ecoflex^{TM} to create the conductive traces and electrodes (rows and columns of the array) [100]. In this case, a conductive threshold was calculated for a CB-PDMS-Ecoflex^{TM} composite with 25% CB, 55% PDMS and 20% Ecoflex^{TM}. As opposed to the prior example, this approach was fully soft but had to be integrated with an Arduino PCB for sensing processing. The device was capable of creating a 3D pressure map of objects. Cataldi and co-workers studied the percolation threshold and elongation ability of carbon nanofibers (CNFs) versus graphene-based elastomeric structures [101]. It was found that due to the longer length of the CNF, the percolation threshold was reached for lower concentrations of material. Besides, CNF were stable even under 100% elongation. As a result, CNF elastomeric interconnects can be useful for stretchable electronics, namely e-skin devices. In a recent study, Zare *et al* proposed a new set of simple equations to calculate the percolation threshold and tunneling distance for nanoparticles. In their study, they focused on polymer-CNT nanocomposites, namely PDMS/MWCNT, ultra-high-molecular-weight polyethylene/MWCNT, poly(vinyl chloride)/MWCNT, PET/MWCNT, and epoxy/SWCNT. From their calculations, they were able to place the percolation threshold of these composites between 0 and 0.06 in terms of effective CNT volume fraction. They also found that the concentration and the dimensions of the CNT were the main effects affecting the percolation threshold [102].

Another technique that can be used to develop stretchable and hysteresis-free connectors for sensors arrays is to develop 3D printable composite pastes that can be cured in a multi-stack arrangement. As an example, Kang *et al* developed three-dimensionally printed pressure sensor arrays by using a 3 axis-programmable dispenser [103]. To develop pastes with enough viscosity to allow for the multi-stack 3D-printing process, they added polystyrene-polyisoprene-polystyrene (SIS) to their paste formulations. Thus, the stretchable electrodes were developed using an Ag flake + SIS formulation, and the piezoresistive sensor pastes consisted of a mixture of SIS with non-destructively amine-functionalized multi-walled carbon nanotubes (NH_2MWNT) and graphene oxides (GOs). SIS insulator was also printed to allow for cross-overs between connectors.

Thanks to their properties, these materials are often used to create stretchable sensors and take advantage of the variations in electrical conductivity caused by stress and strain deformation. For this purpose, origami/kirigami structures and auxetic-inspired designs are also often adopted [98, 99]. These latter designs are capable of bidirectional stretching without cracking, thanks to their negative Poisson's ratio. As a result, they are efficiently integrated over curved surfaces prone to stretching and are widely implemented in wearable and on-skin devices [98, 104].

2.3.2. Use of intrinsically stretchable conductive inks

Finally, another option is to resort to inherently stretchable materials, that rely on conductive formulations based upon the dispersion of metallic particles, or intrinsically stretchable polymers [81, 83, 94]. These inherently stretchable materials circumvent the need to use constituents with different stretching and flexural moduli, as the ones presented above. Such formulations include eutectic indium gallium (eGaIn) alloys, silver-based solutions, and stretchable polymers [70, 105–108]. Among the stretchable polymeric conductors and semi-conductors, the most employed in literature are poly(3-hexylthiophene-2,5-diyl) (P3HT), Polyaniline (PANI), Polypyrrole, and poly(3, 4-ethylenedioxythiophene)- poly(styrenesulfonate) (PEDOT:PSS), whose viscosity can be modified to render them more compliant with stretching and less brittle [83, 109]. As an example, Byun *et al* developed PCB-less stretchable circuits using PDMS as the substrate, which was pre-strained ($\varepsilon_x = \varepsilon_y = 30\%$) during the assembly of the interconnects and SMD (passive components, IC, antenna, radio frequency (RF) module) [82]. In this case, the ink used was a stretchable ink-jettable Ag formulation, and the SMD were integrated using Ag epoxy.

In another practical example, Nagels *et al* proposed the fabrication of multilayer sensing devices [104]. The circuit layer was created using AgNP IJP printed interconnects on top of drop cast PDMS. The component layer was created above the circuit layer and Cu pads were used to integrate the SMD components. Once again and the two layers were connected through vertical interconnect accesses (VIAs), which in this case were filled with Gallistan.

The use of liquid metal alloys is becoming a major issue in what concerns the development of SCE. Their attractiveness rests on the fact they can be stretched to several times their original length and recover their original geometry at room temperature without any associated hysteresis or brittleness [110]. Moreover, thanks to their low melting point, these materials have healing properties, granting that even if a loss of conductivity occurs, it can be restored by applying temperature. For instance, the melting point of gallium (Ga) is 29.8 °C, and adding it to the formulation of liquid metals drastically decreases their melting point. One example is the mixture of indium and gallium in their eutectic proportion (eGaIn), where a melting point of 15.5 °C is achieved. For instance, Nagels and colleagues developed stretchable interconnects by simply injecting eGaIn into VIA generated on a silicone substrate by resorting to a laser-cut vinyl mask [104]. Liu *et al* also developed highly stretchable multilayer circuits using eGaIn. Exposing eGaIn to oxygen leads it to segregate into nanoparticles separated by a gallium oxide layer, which helps it adhere to surfaces, however, it also turns it non-conductive. To obtain highly conductive eGaIn layers, the nanoparticles need to be sintered together which can be achieved by thermal sintering. In their approach, Liu and colleagues deposited the eGaIn layers onto a silicon wafer through spray coating and then heated them in a furnace at 900 °C. After sintering, the eGaIn layers were transferred to silicon elastomer substrates. Extreme stretchability (>1000%) and cyclic stability were achieved and, for proof of concept, the authors developed amplifier circuits, LED arrays, and complex PCB with SMD using this process. Alternatively, Virone *et al* developed large area human-machine interfaces (HMIs) by spray printing eGaIn directly over PDMS using a laser-cut stencil [111]. To achieve a capacitive sensitive matrix, three insulating polymer layers were deposited, intercalated by two eGaIn liquid metal layers patterned in the shape of a mesh.

2.4. Structural electronics

Structural electronics allow for the integration of electronics into three-dimensional structures. These structures are ideally made from light, thin, and durable materials and enhance the functionality of devices, thereby improving human-machine interfaces (HMI) [21].

2.4.1. 3D-MID

3D-MID is a manufacturing concept that allows the development of complex 3D-shaped electronic devices, without the need for conventional wiring processes and FR4 substrate that characterize the traditional rigid PCB [112]. Such devices are mainly obtained by resorting to laser direct structuring (LDS®) technology [113], which emerged in the late 90s and revolutionized the way electronic devices were manufactured [114]. LDS was patented by a well-known German company, named LPKF, and is currently one of the most appraised technologies to develop robust electronic devices, being employed in the serial production of 3D-MID since 2006 [114]. Other technologies resort to two-component injection molding followed by selective metal plating processes, and hot stamping. Even though 3D-MID devices can also be obtained through soft lithography methods, such as 3D soft and holographic lithography [34], direct PE techniques [115], and fused deposition modeling (FDM), LDS is the lead technology for circuit manufacturing onto 3D structures. The process itself encompasses three main critical steps:

(a) The injection molding of a thermoplastic with the desired 3D shape;
(b) The lasering step, which prepares the surface for metal deposition. This step creates crates on the surface of the structure, allowing the posterior adhesion of metal on the thermoplastic polymer during the final process;
(c) The platting step that can be executed through electroless [71], or electrolytic plating [96].

LDS process uses special additive polymers that are activated by the laser to promote the deposition/adhesion of the metallic ink (e.g. Cu, Ni, Au, Ag). Bachy *et al* have recently extended the use of ceramic-based materials to create 3D-MID devices using LDS [116]. In their work, they developed structures based upon alumina ceramic composites with copper oxide additives and studied the influence of the laser process parameters (laser power, velocity, and frequency) on the quality of the final 3D-MID structures.

In recent years, 3D-MID has experienced significant growth, mainly due to the ongoing digital transition that demands the development of increasingly powerful, thinner, and smaller devices. Simultaneously, with the implementation of the Industry 5.0, 3D-MID has established itself as the most adequate solution to rapidly design and prototype robust SCP, encompassing sensors, actuators, and an IoT communication protocol [117]. Faced with the fast-paced evolution that characterizes this technological field, and the need to customize production and develop 3D-MID components in smaller quantities, researchers and industries have recently become invested in alternative routes to obtain these devices [117]. Hence, selective laser sintering, FDM, and stereolithography are being increasingly used, since they bypass the need to create expensive molds for the injection step, which characterizes the first LDS step. Another distinct route to obtaining 3D-MID devices respects laser-induced metallization (LIM) [118]. In this case, the pattern of the conductors is defined by using selective laser irradiation to induce a catalyst deposited over a thin layer of previously coated polymer. Afterward, electroless copper plating is conducted and the patterns are metalized. This technique is widely used to develop QR code stickers and has also been applied to the manufacturing of diverse range of antennas, and even humanoid fingers [118].

To complete the process of device fabrication, SMD components are usually necessary to bring functionality to the structures, hence, PnP technology is frequently associated with LDS and LIM processes [119]. As a result, in industrial manufacturing settings, it is beneficial to combine the 3D-MID structuring technologies with PnP solutions, capable of assuring precise alignment between the 3D structures and the electronic components assembly.

In terms of applications, the 3D-MID devices have been mostly applied in the development of SCP devices for retrofit applications [116, 117]. As previously stated, they are often employed to add control features to machinery in industrial settings and have been crucial to the implementation of the Industrial Internet of Things (IIoTs) paradigm. Another prolific application is the use in the manufacture of automobile cruise controllers [112]. In the latter case, the use of low-cost materials, such as polymers and CNT/polymers composites is a promising alternative for promoting a fast and sustainable production [112, 120].

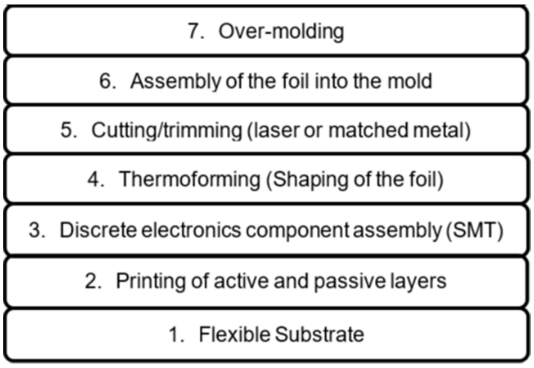


Figure 10. IME process flow.

2.4.2. IME related technologies

Other pathways to obtain structural electronic devices include IME technologies, adopting similar methods to more conventional In-Mold-Decoration (IMD) or In-Mold-Labelling (IML) techniques [121]. FHE and SCE can be combined into structural electronic 3D devices through a series of processes. An IME product usually involves the injection molding of a thermoplastic integrated with electronics [115]. To achieve this, it is necessary to merge the printed and flexible functional foil (from flexible PE), with added traditional PnP components, using in-mold assembly technologies. To finalize the device and improve the user's experience, a decorative or graphical layer is often added. This assembly process can occur simultaneously during the injection molding of the thermoplastic, respecting IMD and IML technologies, or can be done at the end of the part molding process. The final products are end-use molded electronic devices with enhanced functionality and performance [61].

This IME base technology was previously designated as Hybrid In-Mold Integration (figure 10) [122], or functional film insert molding. This process depends on the assembly of a printed and hybrid stretchable electronic layer into a mold (steps 1–3, figure 10) and culminates in a step called over-molding of a polymer. For printing the passive electronics, screen-printing is the most frequently employed technique, as screen printing pastes can be easily engineered to allow for stretching and thermoforming [123]. After printing and functionalization of the foil with PnP of components using conductive adhesives, the foil is subjected to a critical thermoforming step, where the 2D foil is dramatically deformed into its 3D final shape (step 4, figure 10) [124]. To successfully conduct this step, computer simulations are usually made to study whether the printed circuitry and discrete components (SMD) will be able to sustain the thermoforming conditions (temperature, pressure, and deformation) or if adjustments need to be made [125]. After thermoforming the foil, the exceeding substrate is trimmed or cut, so that the entire shape fits the mold, without leaving any extra hanging material (step 5, figure 10). Then, in the over-molding step, the thermoformed layer and components are sealed onto the final structure, simultaneously to its conformation, through injection molding of a thermoplastic polymer (steps 6–7, figure 10). Since relatively high temperatures and pressure are demanded, the embedded circuitry and IC chips should be previously encapsulated [122]. Finally, a graphic inked foil of stretchable and heat-resistant material can be added to the structure to include labels and convey a more functional design to the molded part. This can sometimes double as a protective layer that shields the device from environmental abrasion and moisture [126].

IME is also a fast-growing technology, which is frequently used in the development of the physical counterparts of SCP. As a result, in line with what is being observed in other manufacturing fields, alternative and lower-cost materials for IME have been gathering attention. For instance, organic binders can be added to the printable inks to add stretchability and enhance performance [123]. IMD and IML are also R2R compatible and specialized industrial machinery, capable of producing high-quality IMD and IML parts, has been developed by companies such as KURZ, in Germany and Nissha, Japan [61].

Recently, Ting *et al* conducted a thorough study on the 2D to 3D thermoforming process and created software that took into account the variables that affect the process [125]. The software also aided in the design and optimization of new circuits by detecting and preventing failures. They further proposed a modified IME process by automatizing the surface-mounting process before executing the over-molding step. The process, named *iMold,* is compatible with 3D CAD design software and vows to improve the feasibility and customizability of IME for high-volume manufacturing settings [125]. The software also allowed for testing the placing of SMD and assessing their behavior during the distortion process they suffer when the materials are thermoformed from 2D to 3D shapes.

IME is already widely employed in areas such as automotive, medical, and everyday electronics and household appliances [61], and several IME companies are emerging and growing. One of the leading companies in this market is TactoTek®, which even holds the trademark of an innovative Injection Molded Structural Electronics (IMSE^{TM}) [121, 127]. These technologies present several advantages against the traditional ones since they allow for a high percentage of weight and thickness reduction when compared to the conventional PCB. For instance, using IMSE^{TM} the perceptual weight and thickness reduction reach 70% and 90%, respectively [127]. For industrial purposes, manufacturing lines are usually composed of machinery that executes all IME steps, which makes it compatible with large-scale production [126].

2.4.3. 3D PE

3D printing uses additive manufacturing technologies to generate a versatile and customizable range of structural devices that can be made from either rigid or soft materials. Thanks to its open concept, this technology is easily keeping pace with the ongoing digital transition and, combined with the 3D printing of electronics, has been responsible for the development of smart prosthetics, HMIs, soft robots, wearables, harvesters, and other sensor-actuator devices [128–133]. 3D printed electronic devices can be achieved through 3D printing techniques that have also been appointed above as alternatives to developing low-cost 3D-MID. Additionally, research efforts are also focused on the development of ways of integrating several technologies into a 3D printing machine that will, in the future, be able to create fully 3D printed seamless electronic devices in a single process step [62].

Hence, fully 3D PE represent the complete transition from the complex and time-consuming conventional paradigm to the entirely printable and customizable one-step manufacturing [35, 37]. This much-anticipated technology is currently approaching its full implementation, nonetheless, some constraints still need to be tackled. The minimum requirements include a multi-material 3D printer and a slicing software capable of generating a hybrid g-code encompassing all the required materials. Since those materials present such different physical properties and require different processing technologies, such hybrid slicing software is not yet available. Current multi-material printing software can control several printheads but are quite limited to fused filament fabrication (FFF) [35]. Thus, to achieve a fully 3D printed circuit there is still a demand for more complex and sophisticated software and algorithms. The core goal is to develop the already existing technologies and develop an improved process capable of translating CAD-to-print files into multi-material g-code with the power to define different materials and integrate symbiotic cooperation of FFF, microdispensing, laser cutting, and UV curing [35].

With these tools, it will soon be possible, not only to fully print sensors and actuators but whole devices. One company that is currently focused on the development of equipment for 3D electronics manufacturing is NanoDimensions. They have developed a complex 3D printer and software, called DragonFly liquid deposition modeling (LDM)^{TM} system, which is capable of simultaneously processing a photocurable dielectric and a conductive Ag nanoparticle ink, by depositing them over multi-layered complex PCB.

The system is used for rapid prototyping of PCB, 3D sensors, and 3D antenna devices, with great results in terms of reliability and reproducibility [134]. Marasco *et al* used this technology to develop a meander-like flexible antenna on a PET substrate [135]. The antenna exhibited good performance and was envisioned to be applied to the Internet of Healthcare Things. Another RF antenna manufactured resorting to this technology was envisioned by the Harrys Corporation and developed for the International Space Station using the NanoDimensions 3D printer. It was designed to operate at 5.2 GHz and integrates an RF amplifier capable of enhancing the operation up to 6 GHz. Other devices already produced by this technology include IoT/Wifi access points, 3D-MID devices, coaxial cables, coils, and inductors [135, 136].

3. Applying PE to SCP

The multidisciplinary and complementary technologies described above fall under the comprehensive term of additive electronics and, in most cases, parts of the devices obtained through additive electronics are printed. Consequently, PE technologies are associated with the development of novel SCP, which are generally thin, lightweight, energy-efficient, robust, flexible, and/or stretchable [137]. Key components for the development of SCP include sensors, actuators, light-emitting devices, harvesters (energetic surfaces), batteries, communication elements, and memory units. In figure 11, the mentioned printable functional elements are represented.

Thanks to recent progress in the field of sensor materials, manufacturing technologies, and integration, their cost is falling and their wireless transmission capacity, as well as their self-sufficiency, are improving [142]. Besides, the wireless synchronization of the devices with remote cloud networking systems enables storage, analysis, and communication of indefinite amounts of data between devices [143]. With additive electronics manufacturing pathways as enabling technologies (particularly PE), new SCP can be produced and, in some cases, connected to the IoT, AI and machine learning, cloud computing, blockchain, and big data. As a result, new markets in the fields of IIoT, smart agriculture, food and packaging, transportation, environmental protection, agriculture, medical care, household appliances, buildings, and cities have surfaced [143]. In the next section, each one of the printable electronic components used to produce SCP will be discussed from the optics of functionality and applications.

3.1. Electronic components

Electronic devices are energy-dependent and rely on the synergistic roles of discrete components, which can be passive or active, depending on their purpose in the electronic circuit. Passive components include resistors, capacitors, inductors, and transistors, which suffer variations in their electrical characteristics with environmental changes (such as pressure, temperature, and humidity) and thus, can be used as sensors [144]. Further details about capacitive and resistive sensors are given in section 3.2.

Both passive and active components have been proved printable through the sequential deposition of conductive, semiconductive, and dielectric materials. Zhang *et al* presented a comprehensive example where they used a low-cost, scalable, and fully additive printing process to create different high-gain amplifiers [145]. To achieve this, they used screen-printing and slot die coating to create the functional elements (namely transistors, resistors, capacitors, and inductors, as well as the interconnects that provided their integration into functional differential amplifiers with competitive performance). Correia and colleagues also developed a functional circuit resorting to IJP to fully print passive elements. They created a planar coil equivalent circuit composed of a resistor, an inductor, and a capacitor [146].

Concerning capacitors, the most recent milestone was the ability to escalate their performance by printing supercapacitors (SCs), which have the potential to be used as reliable, clean, and sustainable power supply modules. The first SC achieved through printing technologies was presented by Kaempgen *et al* in 2009 [147, 148]. In this work, SWCNT were used as both conductive electrodes and charge collectors. This group, led by George Gruner, had already been able to print millimeter-thick batteries two years prior [149]. Currently, it is already possible to obtain SC by resorting to a wide variety of materials and employing almost every printing method. Both inorganic materials (carbon-based, metal-oxides, and other 2D materials) and conducting polymers can be used as conductors. Electrolytes allow charge transportation and greatly influence the performance of the SC and batteries. These can be aqueous, organic, or ionic liquid-based gel polymers. Finally, the current collectors (where the charge transporter polymers are attached) can be metal (stainless steel, nanogold, or nano-silver meshes) or carbon-based (usually SWCNT) [150].

Transistors are used in every integrated circuit and can also serve as sensors and switches [151]. Due to their relevance in electronics, they can be classified as the key enablers of the digital transformation and are massively present in modern technologies and devices. As a result, one of the most praised achievements of the PE industry was the development of the first printed thin-film transistor (TFT). Transistors encompass gate and source-drain electrodes, a dielectric spacer, and a semiconductor, which is the top layer of the TFT stacking [152]. Arias *et al* first proved TFT could be fully jet printed and used an organic polymer (poly[5,5′-bis(3-dodecyl-2-thienyl)-2,2′-bithiophene, PQT-12) as the

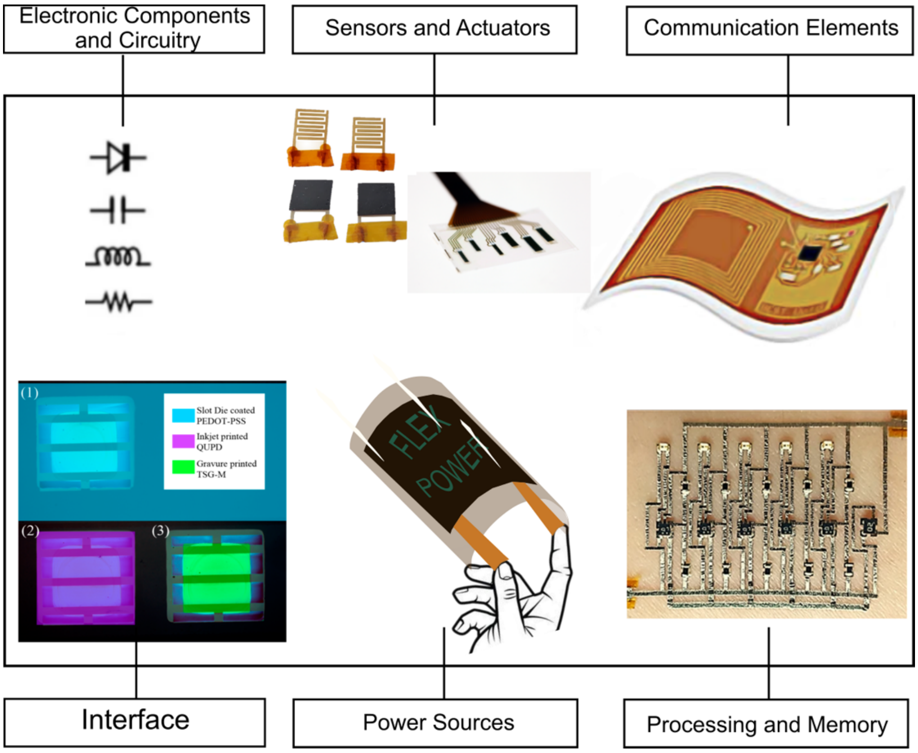


Figure 11. Printable Components used in the development of SCP. Printed piezoelectric polymer actuator reproduced with permission from [138], and piezoresistive electrodes screen printed at Minho University; printed displays reproduced from [139]. CC BY 4.0.; printed communication elements reproduced from [140]. CC BY 4.0.; Illustration of a printed battery; printed temperature monitoring circuit with integrated SMD, reproduced from [141]. CC BY 4.0.

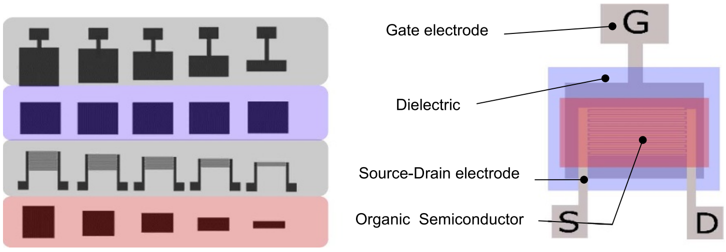


Figure 12. Representation of the successively deposited layers to create a TFT and dimensional ratio variations of the active part. Reproduced from [158]. CC BY 4.0.

semiconductor [153]. Four years later, Fortunato and her group received worldwide recognition for successfully printing the first field-effect transistor (FET) on paper [154]. To create such a device, a transparent conductive oxide was used as the gate electrode on one of the sides of a cellulose sheet, which served as the dielectric. On the opposite side, a layer of an active oxide semiconductor was deposited, acting as the channel gate electrode, conductive drain, and source electrode. This work was the predecessor to the development of fully transparent screens [155].

As frequently mentioned, the advantages of conductive organic materials to formulate ink solutions and pastes have intensified the research and development efforts in this field. Henceforth, many approaches to producing fully printed organic thin-film transistors (OTFTs) [151, 156], and organic photodetectors (OPDs) have been advanced recently [157]. Moreover, printed flexible OTFT can be combined with OPD to create conformable sensor arrays. Sowade *et al* used an all-IJP approach to up-scale the production of OTFT [151]. The group conducted a layer-by-layer deposition of materials in the order depicted in figure 12, and created large-area arrays of 154 TFT each. By varying the dimensions of each component, they conducted a thorough study on the performance of TFT with different channel width to length ratios (W/L).

Thanks to PE many other examples of printed electronic components have been flooding electronics-related scientific journals, confirming the

Table 3. Summary of the core electronic components, their inherit printability and representation in schematic circuits.

	Component	Printability	Illustration		Component	Printability	Illustration
Passive Elements	Resistors	Yes [144–146]		Active Elements	Transistors (OTFT, OFET)	Yes [151–156]	 PNP NPN
	Capacitors and Super-capacitors	Yes [144–148, 150, 164–166]			Diodes	Yes [167, 168]	
	Inductor	Yes [162]			Optoelectronic devices	Yes	
	Multiplexers	Not yet			Amplifier	Yes [169, 170]	
	Antennas	Yes [129, 171, 172]			Integrated Circuits	Yes (CMOS) [173] ARM [174]	
	Power supply	Yes [148, 149, 152, 175]	 (DC voltage) (AC Voltage)		Transistors (OTFT, OFET)	Yes [151–156]	 PNP NPN

growing interest and advances related to these promising technologies [145, 159–162]. By combining these elements, it is already possible to produce complex circuitry capable of processing, such as amplifiers and filters (as approached in section 3.6, however, SiF and SoP CSP technologies are still demanded to integrate SMD. The presence of SMD, such as integrated circuit chips, microcontroller units (MCUs), ARM processors, and analog front ends (AFEs) enables more powerful operations [163]. Consequently, while components such as most of the MCU, AFE, microprocessors, IC chips, and multiplexers, are still not fully printable, alternative designs, thin-film integrating technologies, and meander interconnectors can be used to efficiently and seamlessly integrate them onto flexible and, in some cases, stretchable devices. In table 3, the core electrical components, their printability, and their schematic are represented.

Even though a wide variety of printable components already exists, the large-scale manufacturing of fully printable marketable devices is still in its early stages. This is due to the higher throughput and performance achievable through the traditional routes, as well as their already well-established presence in the market, standardized production, and defined ethical and safety guidelines.

3.2. Sensors and actuators

Sensors and actuators bridge the physical and cyber worlds of SCP. While sensors receive inputs from the physical world and translate them into quantifiable units, an actuator receives that information and responds accordingly [176]. They are integrated into electronic circuits through the adequate use of electronic components that allow for the acquisition and modulation of electrical signals prior to and post sensing.

When employing PE, sensors, and actuators usually present large dimensions (above the micrometer) and can be arranged in large arrays that can measure from a few centimeters to some meters [177, 178]. Oppositely, in microelectronics, sensors and actuators are often classified as micro-electro-mechanical systems (MEMSs), which refers to their micro-meter to sub-millimeter dimensions. Whilst MEMS are mostly obtained through high precision traditional electronic manufacturing methods (e.g. deposition, lithographic, and etching processes), thanks to the fast development and increasing resolution of the PE technologies, printed MEMS and large arrays of printed MEMS are emerging [179].

3.2.1. Pressure sensors

Pressure sensors make up a considerable piece of the sensors market, and their value is estimated to reach $20.8 billion by 2025 [180]. Since these sensors are usually envisioned to be integrated over surfaces, there is a great demand for them to be printable, flexible, and conformable. Besides, they are easily manufactured and can be produced by resorting to simple techniques, and affordable materials. They are generally very sensitive to pressure fluctuations and, as a result, can be used in the manufacturing of ultrasound and vibration detection

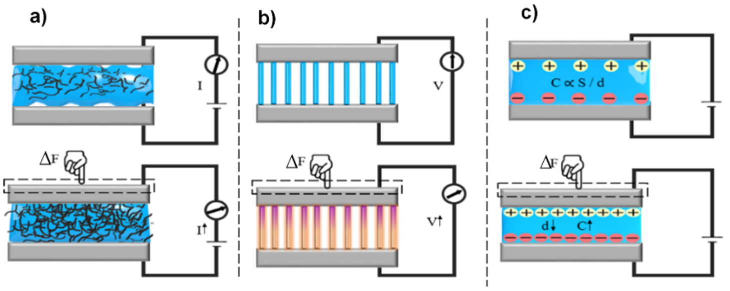


Figure 13. Depiction of the working principle of pressure sensors. (a) Piezoresistive pressure sensors; (b) Piezoelectric pressure sensors; (c) Capacitive pressure sensors (d = distance between conductive electrodes; C = Capacitance value).

devices and haptic touch controllers and displays [181]. Even though these sensors can be produced as individual parts, they are frequently manufactured in the form of large arrays, for large area pressure mapping [182]. Pressure sensors can be piezoresistive (figure 13(a)), piezoelectric (figure 13(b)), or capacitive (figure 13(c)). As previously mentioned, transistors, specifically the FET can also be used as pressure sensors and can be printed in large arrays [36, 183].

Piezoresistive sensors translate external forces into resistance variation that can be detected by changes in the electrical signals [184]. As depicted in figure 13(a)), the application of a force changes the electrical resistance of the strain-sensing percolation matrix (the conductive path is pressed together and the overall resistance decreases) [185], which are then read by the neighboring circuit and can be displayed as a value of force. This low-cost printable technology is also vastly employed to produce strain gauge sensors that have been reported to perform better than the traditional strain gauges [185]. These sensors are usually produced by resorting to composite materials that include a flexible polymer matrix filled with conductive carbon materials including CNFs [186], CNTs [185], graphene [187], CB [188], or conductive metallic materials such AgNPs [189], AgNWs [190], or gold nanowires [191].

Piezoelectric sensors are manufactured using special materials that generate electrical charges when under dynamic forces. Those materials rely on the inversion of their electrical polarization (electrical dipole moment) when under mechanical stress, which is transduced by an increased voltage reading in the circuit. Materials with a high piezo-electrical coefficient, such as ferroelectric polyvinylidene fluoride (PVDF)-based polymers, are usually chosen as sensing materials since they present a high energy conversion ratio and present a fast response [192–194]. Moreover, PVDF-co-trifluoroethylene (TrFE), in particular, also exhibits pyro-electrical response and can therefore be used as a thermal sensor [195–197]. As a consequence of their high sensitivity to pressure fluctuations, piezoelectric sensors can also be used as acoustic sensors and resonators for the production of traditional loudspeakers [169], or to create miniature-sized sensors intended to be applied in the production of electronic stethoscopes [198], or even implantable cochlear implants [199]. Another interesting property of these sensors is the capability of functioning as energy harvesters. By taking advantage of vibrations (from human movement, machines, pumps, vehicles, railway tracks or wagons, floors, walls, etc) these devices can be optimized to work as self-powered systems [200, 201].

The capacitive sensors can also be used as pressure and strain transducers by converting geometry variations between parallel plate electrodes into capacitance changes. Capacitive sensors are very effective for large-area electronics and are frequently used in the form of arrays for pressure mapping and multi-touch applications [202]. Due to their sensibility, they are also efficient proximity sensors, which means they can detect the presence of nearby objects without direct physical contact [203].

To sum up, pressure sensors, are some of the most ubiquitously distributed sensors in modern society. They are applied to sectors such as industry, automotive, healthcare, and entertainment. Combined with AI they can be applied in areas such as object recognition [100, 204, 205], object handling with pressure control features, industrial PnP [206], weight and shape characterization [207, 208], soft prosthetics, electronic skins [83, 194, 205], haptic feedback [204, 205, 209], and robotic grippers [210, 211]. One of the leading companies in printed pressure sensor technology is Tekscan, which produces pressure mapping, force measurement, and tactile sensors as well [212]. These sensors have been patented as the F-Scan^{TM} system (that uses FlexiForce^{TM} sensors) and are used in sports for monitoring athletes' performance (e.g. in-shoe sole sensors) and in orthodontics, for image reconstruction of the dental morphology of patients. Tekscan technology has even been also used in animal research, to monitor animal activity (including movement dynamics and orthopedic disease recovery) [212]. Piezotech is another leading company that specializes in the production of fluorinated electroactive polymers (mostly PVDF-based polymers) and their products include pressure, touch, and vibration sensors and energy harvesting solutions based on printed piezo- and triboelectric nanogenerators [138].

3.2.2. Environmental and electrochemical sensors

Environmental sensors include temperature, relative humidity, and chemical element transducers. These sensors have been extensively studied and there are countless examples of these devices. They can be produced on a large scale, and they are usually low-cost, and exhibit low-energy consumption rates that make them able to function remotely for long periods. Environmental sensors are particularly important for climate and air quality control in smart homes and smart cities [213], or to monitor biophysical signals of patients [214–216]. Temperature sensors are manufactured by resorting to functional conductive materials that change their resistivity with temperature fluctuations and cause variations in the current flow of the circuit [214]. This variation is usually stable and presents good linearity, which indicates these sensors' reliability, even after several measuring cycles. The resistance–temperature relationship can be positive or negative. In the case of a positive relationship, the sensors can be either resistive temperature detectors (RTD), or made from positive thermal coefficient (PTC) materials [79]. When the relationship is negative, the used materials are known as negative temperature coefficient (NTC) ones. Both inorganic (e.g. silver inks [214]) and organic materials (e.g. polymers such as PEDOT:PSS [217], or PVDF-TrFE [195]) can be used in the manufacturing of these sensors. The electrodes of these sensor devices are frequently printed in a planar meander-like or interdigitated arrangements and can be printed in arrays to create large-area sensing surfaces [177]. Recently, Zubkova *et al* integrated temperature sensors into arrays for large-scale applications [218]. Even though temperature sensor arrays are not unheard of, this approach stands out for the materials and technologies selected. The fully printed arrays were made from screen-printed Ag electrodes. The active material was made from a jettable MWCNT (0.05 wt%) ink, functionalized with carboxyl acid (–COOH) and the substrate was a flexible PI sheet. To encapsulate the active material, a UV curable ink was deposited on top of it. To allow for localized and sensitive data collection, while simultaneously suppressing cross-talk a zero potential circuit was used and the interconnections between sensing units were divided between the inner and outer layer of the PI sheet, which served as a dielectric and allowed cross-overs between connectors. Thermocouples have also been successfully printed [219]. In a work by Knoll *et al*, a thermocouple array was obtained by screen printing CB and silver pastes on a PET foil [220].

Humidity sensors are also of great importance in several fields from agriculture, food production, and transportation to healthcare. These sensors should exclusively and accurately quantify environmental humidity and be stable to thermal variation, changing their electrical resistance only according to the environmental H_2O concentration [221]. These sensors can be printed and, their readout can be measured as resistance or capacitance variation. Their design usually incorporates interdigitated electrodes (IDEs). To produce these sensors an active film (material that absorbs water molecules) is added on the top of the conductive electrodes through deposition or printing techniques. In some cases, the substrate itself can be the active film [222]. Regarding the functional materials, some examples include the use of high-water absorption films such as titanium dioxide (TiO_2) nanoparticles and MWCNT films. As an example, Ghahremanpour *et al* used spray printing to create humidity-sensitive thin films of MWCNT dispersed on polyvinylpyrrolidone (PVP) [221]. PVP is a hygroscopic polymer that swells during the uptake of water vapor. This swelling disturbs the conductivity established by the percolation path of the MWCNT, and by studying its percolating behavior, the specific volume resistivity of the material can be correlated with quantifiable humidity concentrations. Similar approaches can be adapted to other materials such as polyvinylalcohol (PVA) [223], PVA/CNT-based sensors [224], and PVA/PEDOT:PSS-based sensors [225]. Recently, Wen *et al* developed smart connected multifunctional clothing made from electrospun PEO and silk fibroin [226]. AgNW electrodes were spray-coated over a silk fiber film through a mask to create the desired IDE geometry. The active solution of Silk/PEO was then electrospun between the electrodes and by measuring capacitance changes it was possible to obtain pressure, temperature, and humidity sensors.

Electrochemical sensors suffer variation in electrical conductivity when in contact with varying concentrations of certain molecules. Consequently, these sensors can be used to detect and quantify the presence of a wide variety of toxic substances, metabolites, or biological markers in various settings. CNT-based sensors, for example, can measure volatile organic compounds (VOCs), as well as other gases, such as oxygen (O_2), carbon monoxide (CO), carbon dioxide (CO_2), nitrogen (N_2), ammonia (NH_3), amongst others [227]. Another example of these sensors are the glucose detectors, which are indispensable for people living with diabetes. Sensors such as these are already achievable through PE and their low cost is an advantage that allows them to be distributed all over the world to everyone in need [228]. 'Electronic noses' or 'eNoses' are also electrochemical sensors that can selectively sense odors and flavors [229]. Such multi-sensory systems must be connected to multiplexers that recover data from multiple sources and cross it with a pattern-recognition system that responds to different odors [230]. These sensors can also help monitor the air, water, and soil quality as well as the presence of highly dangerous chemicals including nerve and pulmonary agents [231]. They have also been applied in the medical field to help diagnose and monitor certain physiological conditions in the human body (e.g. by detecting VOC in exhaled breath) [229].

Since more often than not environmental and electrochemical sensors need to be sending data remotely, they are usually paired with RFID or NFC tags [232]. Due to their simple implementation and easy scale-up, their utility is spreading to farming monitoring, food quality control, and logistics.

3.2.3. Photodetectors and optical sensors

Photodetectors include photoelectrical sensors and photovoltaic cells. Light detectors can also be used to detect light frequencies other than the ones from the visible spectrum. One example was proposed by Figueira *et al* that produced UV light sensors using cork as a substrate [233]. For this, the group used a ZnO ink vehicle prepared by dissolving 5 wt. % of ethyl cellulose in an 80:20 toluene/ethanol solution. The ZnO/EC layers were screen-printed over a cork substrate, and IDE electrodes made from carbon ink were also screen-printed over the active layer. Under UV light irradiation, electron–hole pairs are photogenerated in the ZnO/EC layer creating a measurable photoresponse. In the medical field, visible and near-infra-red (NIR) photodetectors are used to monitor biosignals, such as heart rate and blood oxygenation saturation (SpO_2) [234]. The light signal penetrates the skin, and the reflected light retrieves information relative to the heartbeat. Photoplethysmographs (PPG) use the reflected light to translate the signals into SpO_2, upon algorithmic processing. OPDs based devices can be used for this purpose [234]. The advantage of these components relates to their printability, flexibility, and large-area compatibility, which enables full-body sensing (which is a valuable application for burn and skin graft monitoring, for example). Their structure encompasses a flexible substrate, a transparent thin-film barrier (TFE), a thin film transistor (TFT) backplane, a solution-processable OPD layer, and an encapsulator.

Optical sensors are often based on infrared light-emitting diodes (LED) coupled with photodetectors and can be used for tactile sensing and proximity sensing. These sensors take advantage of the optical light reflection that occurs between different materials and extract and process that data into valuable information [235]. Although most of them are made from a simple LED, some applications already incorporate organic light-emiting diodes (OLED) instead [236]. OLED encompass a conductive and emissive layer made from organic molecules or polymers that are inserted between an anode and a cathode electrode. These organic emitting devices have also paved their way as displays that incorporate organic photodiodes for fingerprint imaging and scanning, which allows the users to easily validate their identification when needed [236].

Photovoltaic panels are another type of photodetector. They are the central piece of the solar industry, which is one of the most significant industries in the field of energy harvesting. Given the interest in improving the methods for sustainable power production, its importance is bound to increase for years to come. As a result, this technology is currently also undergoing a transition respecting the preferred functional materials employed in their manufacturing. Since this market is so significant in terms of manufacturing output, it is a priority that non-sustainable materials and technologies are replaced by lighter, flexible, sustainable, and recyclable-friendly alternatives. As a result, a pronounced increase in research regarding thin-film photovoltaics including perovskite solar cells, organic solar cells (OSC) [237], and quantum dot cells (QDC) has been registered, growing 16.4% in 2020 alone [238]. OSC large-area panels can be obtained through large-scale fabrication using R2R production (by printing or coating the active layers and subsequent laminating the completed modules) [237]. These technologies are used to produce large, lightweight, low-cost, and flexible solar panels whose power conversion efficiency is expected to soon surpass 15% [239]. It is also noteworthy that large-area devices experience PCE losses from higher resistance within the OSC, and losses related to the higher ratio of dead areas that arise from the geometry of the larger area panels (as a result, smaller devices present higher PCE). Another technology on the rise as PV cells are the QDC which usually use lead sulfide [240], lead selenide (PbSe) [241], or lead telluride quantum dots in the form of colloidal solids [242]. Even though all these materials are low-cost and of easy formulation, Hu *et al* have proven that PbSe QDC enable higher charge carrier generation than the other lead counterparts, reaching efficiencies above the 10% [241]. Nonetheless, the most promising competitors are the perovskite solar cells, which are now emerging as possible future market leaders. The PCE for the perovskite solar cells currently reaches about 23.3% in laboratory settings [243]. Another application of perovskite relates to their application as high-energy flexible radiation detectors. By dissolving 2D perovskite single crystals in an organic solvent (anhydrous dimethylformamide) Tsai *et al* produced large area, 2D layered perovskite x-ray detectors [244].

One of the most prominent companies developing perovskite alternatives to traditional PV is Saule Technologies [245]. This company was the pioneer in the IJP of perovskite solar cells for the production of flexible, lightweight, ultrathin, and semi-transparent photovoltaic modules and is expected to maintain a steady growth in the years to come [243, 245]. Another key feature that differentiates Saule's perovskite technology is their purpose to commercialize a product that is already intended to be connected to the IoT. For this purpose, their solar modules are wirelessly connected to devices that help in their configuration, power management, and energy storage efficiency. As a result, this innovative technology vows to deliver a longer device lifetime, improved transmission range, and security, amongst other advantages.

3.2.4. Thermal actuators

The interest in thermoelectric (TE) devices has been gradually increasing in the past few years. TE devices can function in heating (thermoelectric heaters, TEH) and cooling (thermoelectric coolers, TEC) modes or can be thermoelectric generators (TEG) [246]. TEC and TEH, in particular, are thermoelectric heat pumps that depend on the Peltier effect to cause temperature variation between the different sides of a semiconductor material (absorb heat on the cold side and release heat on the hot side) [246]. In this case, the heat transfer is dependent upon electrical energy consumption. Some areas that use TEC and TEH include industrial, aerospace, automotive, and the military [246].

Due to the popularity of TE devices and their relevance for applications in SCP for remote heating and cooling control, organic-based thermoelectric materials are increasingly desired [247]. Conjugated polymers (e.g. PEDOT:PSS, P3HT, PANi:CSA), coordinate polymers (e.g.: ligands 4,4′-dihydroxy-3,3′-diacetyl biphenyl bisthiosemicarbazone and 4,4′-dihydroxy- 3,3′-dipropionyl biphenyl bisthiosemicarbazone), and small molecules (e.g. pentacene, C_{60}, and thiophene derivatives) can be highlighted as materials capable of acting as thermoelectric devices. Printed thermal actuators usually are applied to heater pad systems, which are already being implemented in car seats, and heated textiles/ wearables [248].

3.2.5. Mechanical actuators

Mechanical actuators such as switches, pumps, and gears need to have a certain degree of mechanical robustness and as a result, are not commonly manufactured using PE. Additive technologies such as 4D printing, however, can already be used to achieve these devices and can incorporate PE [249]. Focusing on PE, even though electroactive inks are already available, their mechanical strength and reliability still need to be improved. Notwithstanding, printing technologies, can already be used to deposit electroactive polymers (EAPs), and other electroactive materials such as ceramic and metal alloys (such as lead zirconium titanate and aluminum nitride) [250, 251]. EAP can be dielectric (more used for low-frequency actuation applications) or ionic materials (used for high-frequency modulation applications) [250]. Regarding dielectric EAP, their actuation is caused by electrostatic forces generated in between the electrodes, surrounding the functional material. The most frequently used ones respect ferroelectric polymers and liquid crystalline polymers. In the literature, the most frequently used ferroelectric materials are polyvinylidene difluoride (PVDF) and PVDF-based polymers, which are piezoelectric. PVDF and PVDF co-polymers such as PVDF-TrFE, chlorofluoroethylene PVDF–TrFE–CFE, and chlorotrifluoroethylene PVDF-TrFE have also been formulated as inks and are commercially available from companies such as Piezotech® and PolyK^{TM} [138, 252].

Another material that can be used as an actuator is PEDOT:PSS, as demonstrated by Byun *et al* [82]. In their work, they were able to remotely activate the fingers of an electronic skin, which were made from a thermoelectrically actuated PEDOT:PSS/PDMS soft bilayer. This actuation occurred due to a controlled Joule heating–induced operation mechanism that caused bending deformation with increasing applied voltages. Several research projects are focusing on these materials and taking advantage of their characteristics, not only as actuators [253], but also as sensors [254], and energy harvesters [255, 256].

3.3. Interfaces

3.3.1. Haptic interfaces

This subsection appears closely related to the one presented above, as many haptic interfaces can be obtained from mechanical actuators. When designing devices or objects intended for human use, it is important that such devices are capable of providing haptic feedback, thus guiding and enhancing the user's experience. Depending on the nature of the device, the haptic feedback can be promoted by a passive structure, as simple as a three-dimensional button, bump, or texturized surface. On the other hand, the device can actively react to the human's touch and shift its properties as a way of delivering information to the user (mechanical actuator) [257]. To achieve this, printed EAP are already being used, as detailed above in the mechanical actuators subsection. In contact situations, haptics can artificially simulate the sense of touch by recreating force and tactile feedback, serving as a strong complement to visual and audio perception. When haptic gloves or on-skin sensors are used in combination with remotely actuated robots or grippers, for example, remote sensing, touch, and grasping operations are enabled. As an example, in a work by Poncet *et al*, an ink formulation of PVDF-TrFE has been used to develop haptic buttons, whose actuation was previously modulated by resorting to the finite element method [253]. The EAP actuator, which can be seen in figure 14, was developed by screen-printing the PVDF-TrFE ink and physical vapor deposition of the gold electrodes on a PC substrate.

Yoon *et al* also developed a sensor/actuator unit resorting, once again to PVDF-TrFe, intended to be

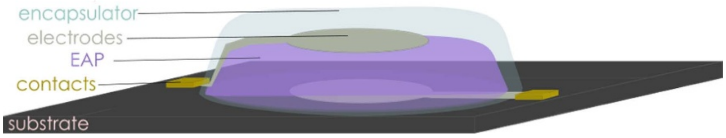


Figure 14. General cross-section schematic of a piezoelectric EAP button.

used for HMI purposes. For this purpose, only printing technologies were employed, and the electrodes were made from screen-printed silver [254]. PVDF–TrFE–CFE is another co-polymer that can be used for low-frequency actuation of surfaces and displays [258].

Haptic interfaces, along with mechanical actuators can be used in prosthetics, to help retrieve the sense of touch in cases where it has been lost or impaired, or in remotely operated robots in settings that include teleoperated medical interventions [259], handling of materials in extreme environmental conditions [260], or aerospace research [261]. They are also vastly employed in the day-to-day and entertainment applications, such as feedback buttons in mobile phones, touchpads, or car dashboards (vibrate as a simple binary cue to inform of on or off states) [262]. Recent technological advances have also allowed the addition of haptics to enhance and support the virtual reality (VR) and augmented reality (AR) experiences [181, 263]. In the particular case of HMIs, the haptic touch has become an integrative part of the information flow that is established between the user and machine, and can be present in the form of graspable, wearable, and touchable systems [257, 262]. Multi-touch displays, for example, encompass touch sensors, controllers, and embedded software drivers that often render them incompatible with thin and bendable applications.

The development of haptic interfaces is expanding at a fast pace and many pre-existent companies, such as Fujitsu [264], and Texas Instruments already commercialize printed haptic-feedback actuated devices. Simultaneously, new companies are also rising to prominence and promise to shift the paradigm of haptic actuation to a thinner, more compliant, and more versatile one. Among them, Hap2U [265], CEA tech [250], Aito [266, 267], and Actronika [268], can be highlighted.

3.3.2. Displays

The simplest optical actuators encompass single or multiple LED that can be turned on or off as digital binary outputs resulting from data processed from sensors [269]. One of the most familiar and omnipresent actuators in our daily lives are the displays, that result from large arrays of LED, and more recently OLED. OLED displays are already an established technology used as television and mobile phone screens, as they allow for an improved energetic performance, color balance, and non-flat conformation when compared to previous technologies (such as plasma screens). These displays are also being used to produce foldable mobile phones and can even be printed over textiles or skin. Another type of display that is fully compatible with printing technologies are the electrochromic displays. These can be based upon PEDOT:PSS, which is a semiconductive and electrochromic material. In its oxidized phase, PEDOT:PSS is gray/transparent, however, when activated by an electrical field, PEDOT:PSS is chemically reduced and its color shifts to blue. Some companies commercializing these types of displays include Ynvisible [270], and the research institute of sweden (RISE) [271].

A leading company in display technology is Samsung, which has recently launched the concept of the first double-folding OLED panel [272]. Similar concepts had previously been proposed by "The Creative Life" (TCL) and "Life's Good" (LG) but are yet to reach the market [272]. The development of flexible displays for the automotive industry is another hot topic being currently disputed by several companies worldwide [273–275]. FlexEnable, for example, has developed the organic LCD technology to face limitations associated with OLED displays [276]. They resort to OTFT materials, branded as FlexiOM^{TM}, and have submitted over 800 patent applications in the last few years [277]. With OLCD technology, large and ultra-bright displays can be achieved with relatively low associated costs [274]. The year 2021 has also seen the upsurge of the first open platform kit for the development of flexible electronics named RoKit. In this kit, users can access a fully flexible micro-LED (mLED)-based flexible display, and a fully flexible sensor module, among other components for software development [278]. They have also recently introduced the world's first mLED stretchable display, capable of 3D free-form shaping, twisting, and convex and concave deformation [279]. Further applications of the displays are their implementation as flexible projectors for VR or AR purposes [145]. With the development of these technologies, a demand for better performance and enhanced resolution quality is needed and, therefore, innovative flexible micro-displays, capable of spatial reality are under intense expansion. Such micro-displays include OLED, Quantum-dot-LED, digital light processing [146], Liquid crystal on silicon [147], OLED-on-Si [148], and mLED displays [149].

3.4. Communication elements

To achieve some of the applications stated in the previous sections some of the SCP might need to be remotely connected. To function properly, these complex and heterogeneous systems require the support of adequate communication networks, called Wireless Sensor Networks (WSNs), and protocols capable of assuring good throughput, low latency, and suitable range [280]. An example was advanced by Farooqui *et al*, who created a large area environmental monitoring system composed of several mobile wireless sensor nodes [281]. The system used ZigBee WSN and communicated through an IJP antenna with three-dimensional geometry. The antenna was powered by a 45 mAh thin lithium-polymer battery and operated at 2.4 GHz through a 1.5 λ_0 dipole. IJP antennas have also already been employed in 5G communications. To achieve this, Ahmad *et al* printed an AgNP antenna on top of Kapton substrates [282]. The conductivity of the demonstrated antenna was optimized by varying its thickness, and a resonant frequency of 27.75 GHz was achieved. This result was proved adequate to communicate data in the 5G applications' band.

Shorter range communication systems rely on RFID technology. This technology is versatile and is employed ubiquitously in devices around us. In the near future, RFID is even expected to replace the Universal Product Code since it presents advantages in terms of automation, inventory control, and data transmission [283]. Depending on the envisioned application and range, RFID devices can be active, passive, or semi-passive. A basic passive RFID system comprises a reader and an RFID tag. The RFID tag must include an inductor and a capacitor that are responsible for producing the resonating frequency. Reader-powered RFID and near-field-communication (NFC) tags can be developed as polymer-metal composites, and are extremely useful as they assure long-term autonomous performance of devices [284]. To make them competitive in terms of price, performance, and reproducibility they are integrated with thinned and encapsulated logic and memory parts and are therefore not fully printed.

Khalid *et al* developed an ultraHigh frequency (UHF) RFID setup, comprising a capacitive sensor, an RFID tag, and an antenna, capable of functioning wirelessly and battery-less [285]. In this case, the RFID tag functioned as the energy harvester, remotely feeding the system by receiving unmodulated RF signals from the reader. This three-port remote sensing system was envisioned for low-cost and autonomous healthcare or environmental control applications. In a recent publication, Machiels and colleagues developed screen printable high frequency (HF) RFID tags and studied their performance on top of over 70 different fiber-based substrates [286]. The RFID tags were intended to be used on packaging for logistics and transportation control. The developed antenna resonated at 13.56 MHz, and was compliant with a series resistance in the range of 20–25 Ω, and inductance in the range of 2.90–3.00 μH. The tag was combined with a microchip enclosing an analog-to-digital converter, and a wireless IC containing a barcode, which was identified by a reader device. Since the core goal of this work was to evaluate the potential of these tags for third-party logistics it was assured that the tags were reusable by re-setting their RFID code. Their recyclability was also studied and no issues were detected.

In a different study, Kumar *et al* conducted a performance analysis on a flexible NFC tag for IoT applications [284]. The NFC tag, which also operated at 13.56 MHz, was tested for two types of interconnect materials (printed copper vs. graphitic carbon). In this test, copper outperformed graphitic carbon because the latter material presented much larger electrical resistance, which impaired the resonant capability of the antenna. Boada *et al* also took advantage of the simplicity of this technology and developed battery-less NFC sensors for pH monitoring [287]. For this system to be feasible, the mentioned tag needed to present a consumption lower than the energy harvested by the NFC IC from the reader. Throughout this work, the researchers proved the tag was able to work at very low power, consuming 300 μA at 3.3 V. Other examples of battery-less RFID devices obtained through additive manufacturing have been flooding the scientific and technological community for their easy manufacture and convenience [135, 288, 289].

3.5. Power

3.5.1. Batteries

For some consumer products and disposable devices, small and non-rechargeable batteries can be used as the power source. However, for the majority of flexible and stretchable active devices, the bulky coin battery cells are disadvantageous and offer resistance to the seamless integration and functionality of the smart devices. Besides, there is a great demand for long-lifetime devices that eventually outlive the endurance of the small batteries [290]. As a way to circumvent the limited lifetime of batteries, researchers sometimes include rechargeable power units in the systems [291].

In what concerns the additive manufacturing of power sources, there has been an increase in research and development concerning flexible and stretchable batteries [175, 292], and SCs able to offer high energy density, power, long lifetime, and be wirelessly rechargeable or self-sufficient [150, 293, 294]. Screen-printing is currently the leading technology to obtain printed batteries and its potential is being explored by specialized companies such as VARTA [295]. As a market leader in this field, VARTA has also acknowledged the need to implement printable technologies to obtain their products and is currently cooperating with research groups to test several electrochemical printable systems for batteries [295]. Both stacked and co-planar battery designs are printable, and serial multi-cell modules can also be printed using R2R screen-printing of current collectors, cathodes, anodes, and electrolytes. Among the different electrolyte systems used in battery manufacturing, Zink/Manganes oxide and dioxide batteries are currently the easiest ones to print since they can be formulated as aqueous solutions, while also being low-cost, and environmentally friendly [296]. Zink/carbon batteries are also an extensively pursued subject, particularly in the development of long-term evolution tags [297]. Recently, several companies have cooperated in the development of the innovative Bayer® smart label, which is powered by a 2 mm thick Zink/ carbon biodegradable battery [298].

3.5.2. Energy harvesting

To allow for power supply with no constraints, the integration of energy harvesting devices such as OPV, RF antennas, thermoelectric (and OTEG), piezoelectric, triboelectric, and biochemical generators is often desired. Besides, these types of powering solutions are clean, sustainable, and low-cost. Some of these energy harvesting devices can even be used without an associated battery. Examples include passive devices [299], applications that demand low-power consumption [300, 301], or cases where an abundant light source is continuously available. Another alternative that has not been mentioned in this review are the thermoelectric generators, which rely on the Seebeck effect and convert temperature gradients into electrical power [247]. These devices are useful to upcycle heat dissipated from electrical equipment and can be employed as energy harvesters in ultra-low power applications [247, 302]. OTEGs can be obtained using the printable materials, and are able to harvest energy from dissipated heat below 200 °C, elevating them as an attractive solution to harvest energy from households and even human bodies. These organic harvesters can then be used to wirelessly power IoT sensor nodes and used to enable low-power communication networks [303]. OTEG are lightweight, printable, and can efficiently replace inorganic ones, presenting inherent flexibility, low thermal conductivity, and being compatible with large-area manufacturing [304]. In table 4 different energy harvesting technologies are summarized along with their respective advantages and disadvantages.

Some application examples from the literature include the work from Marasco *et al* that produced a flexible antenna for harvesting energy from surface acoustic waves and power sensors [135]. In this case, the vibrational energy was harvested by piezoelectric resonators and used to power an antenna, which was capable of receiving and transmitting information in the frequency band of 2.4 GHz. In another innovative and sustainable approach, Ferreira *et al* proposed a touch-interactive flexible smart card, which exhibited power and current density of 1.75 $\mathrm{Wm^{-2}}$ and 33.5 $\mathrm{mAm^{-2}}$ respectively [309]. This device was intended for IoT applications and was produced by screen printing electrodes made from carbon and silver. The energy-generating (functional) layer was composed of filter paper functionalized with conductive PANi nanostructures (pfc-paper), and the energy was generated by pressure strains derived from motion or touch-interaction, which caused charge transfer to accumulate on the charge collector layer. Finally, to obtain OTEGs, the most popular conjugated polymer is the PEDOT:PSS, which can be spin-coated, drop-casted, inkjet, or screen printed over flexible substrates with high thermoelectric performance results [303]. Recently, Zhu *et al* have started exploring the potential of moisture-induced electricity generation [308]. In this work, they used modified oxygen-based groups of graphene to achieve GO/PVA moisture-electric generators. These generators work by absorbing water molecules and releasing ions, generating separated charges for electric generation. The manufacturing process consisted of blade coating a substrate (FTO glass) with the GO/PVA solution, and drying to obtain a functional film. An Ag paste was then coated on top of the film to obtain the top electrode.

3.6. Processing circuitry and memory devices

As detailed in section 3.1, it is possible to fully print logical processing circuits by studying and implementing several printed passive electronic components in association with flexible printed interconnects. Chang *et al* fully screen-printed high gain amplifiers and digital-analogic converters by optimizing the printing of transistors, resistors, capacitors, and inductors [145]. More recently, Sekine and colleagues successfully integrated COTS and flexible silver printed interconnects to low-operating voltage pressure sensors, which were paired with an analogic amplifier circuit [170]. The schematic design of the device can be seen in figure 15. The pressure sensor was piezoelectric and made from screen-printed PVDF-TrFE and the electrodes from PEDOT:PSS. The source and drain electrodes of the amplifier circuit were inkjet-printed using AgNP ink, and the dielectric parylene was added by CVD. The circuit was finally encapsulated using spin-coated cross-linked poly(4-vinylphenol) (P4VP).

When complex computation is needed, data processing units and memories are often demanded. Microprocessors are semiconductor devices that usually rely on rigid MOSFET fabricated on crystalline silicon wafers [174]. Recently, however, Biggs *et al* have successfully developed a plastic-based flexible ARM microprocessor using indium–gallium–zinc oxide TFT etched on PI substrates. The plastic

Table 4. Comparison of different energy harvesting devices. [290] John Wiley & Sons. © 2016 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

	Energy Source	Advantages	Disadvantages	Estimated power scavenged per cm^2	References
PV solar cells	Light	High power	Not always available; dependent on surface orientation	100 mW (outdoors) <500 μW (indoors)	[305]
Antenna	RF energy	Reliable and versatile (transmission and communication)	Low power	<1 μW	[285, 287–289]
Thermo-electric	Heat (from surface or body)	Reliable and allows for self-powered temperature sensing	Difficulty in detecting small thermal gradients	60 μW	[305]
Piezoelectric	Motion from device or body	High power (for some motions); self-powered motion sensing	Variability of vibrations and movements	200 μW	[135, 305, 306]
Triboelectric	Motion from device or body				
Biochemical	Oxidized lactate, glucose, ascorbate, moisture	Self-powered biochemical sensors	Low concentration variability	<350 μW	[307, 308]

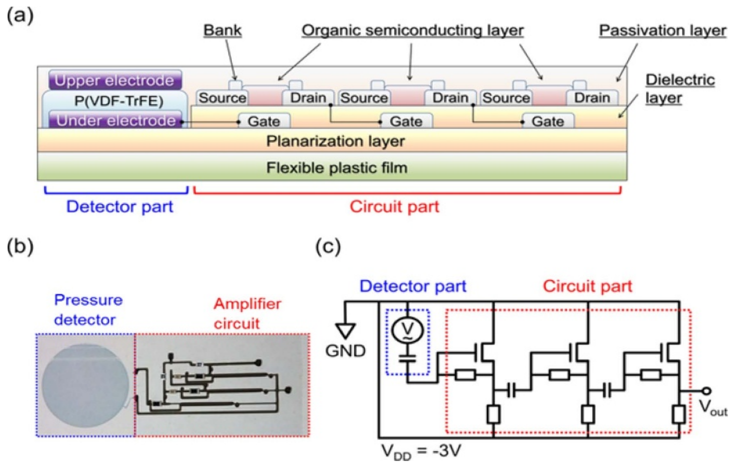


Figure 15. (a) Fabrication steps and device structure of the printed pressure sensor (b) Photograph of the printed pressure sensor in the flexible state. (c) Schematic circuit diagram of the sensor. Reprinted with permission from [170]. Copyright (2019) American Chemical Society.

ARM was implemented with PragmaticIC®, meaning that considerable steps towards fully additively manufactured processors are already being taken.

Printed memories are also already a reality and usually appear paired with logical processing circuits, as exemplified by several works throughout the literature. In 2011, the Norwegian company Thinfilm, along with PARC, launched the Thinfilm Memory^{TM}, which became the first commercializable non-volatile memory device with complementary organic circuits [310]. In 2021, Ramon and colleagues developed simple write-once-read-many (WORM) memories using IJP [16]. To obtain these memories they used the rapid electrical sintering method, which was compatible with the large-scale fabrication of these memory devices. NFC and RFID tags can also be used to store and communicate simple logical information. For this purpose, Le *et al* integrated discrete logical, memory, and battery components with an IJP antenna and interconnects to produce an active RFID-based wireless sensor, which functioned in the high-frequency range [311]. The device, a wireless gas-sensor, included an active RFID antenna, which was successfully printed resorting to an environmentally friendly aqueous solution of SWCNT and a paper substrate. In the same work, they also explored the potential of the same active materials for passive RFID. With that in mind, they created a passive and battery-less solar-powered tag to be applied in health monitoring with integrated discrete MCU components, which communicated wirelessly at 904.4 MHz. In both approaches, the authors were able to achieve

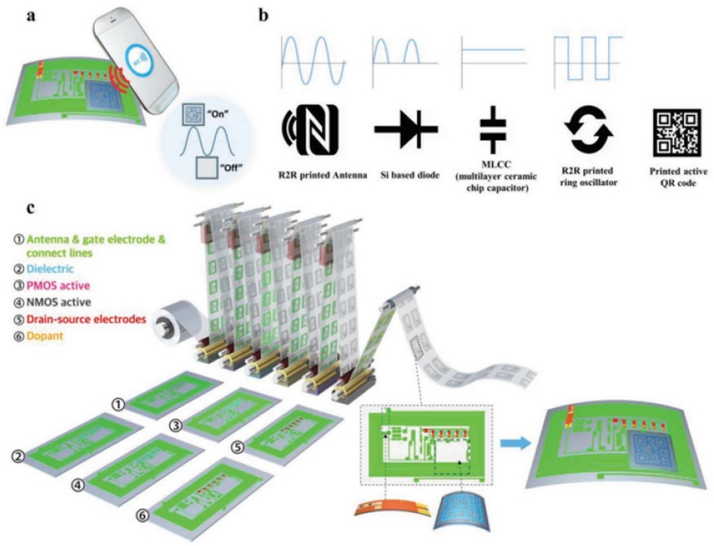


Figure 16. (a) Descriptive illustration for the working concept of NFC QR code label, (b) functions of each device unit, and (c) proposed R2R gravure printing sequences to print layer by layer to complete NFC QR code label. [312] John Wiley & Sons. © 2019 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

good overall performances, nonetheless, they noted that some challenges, respecting long-term accuracy and reliability, still need to be tackled.

In a work by Sun *et al*, a wireless 1-bit-code generator was gravure printed along with an electrophoretic QR code [312]. The circuit consisted of a printed NFC antenna (Ag nanoparticle ink), which was connected to a printed complementary metal-oxide-semiconductor (CMOS)-based ring oscillator (using silver ink, semiconducting ink, and n-doping ink). The electrophoretic QR code was developed using an E-ink, which was printed and laminated over PDMS, and activated upon application of an electric field. The electrodes were printed resorting to blade-coated silver ink and, to create the processing circuit a Si-based voltage divider (made from COTS: two diodes and two capacitors), and an R2R Gravure Printed Buffer Inverter were integrated. The latter was prepared by printing a p-type TFT and a resistor. The manufacturing scheme of this NFC device can be seen in figure 16. By integrating the QR code with the wireless 1-bit code generator this NFC device was able to act as a WORM memory and carry 2953 bytes of information by functioning in combination with a smartphone and camera.

4. Concluding remarks

SCP along with the IoT-related concepts are expected to continue to expand and connect devices, services, people, and infrastructures. Increasingly simple solutions are prone to emerge for various markets with lower associated costs and higher sustainability. This is powered by research in multiple fields, from materials science and electronics, communications, data science, and hardware and software development, among others. The large-scale implementation of PE technologies, in particular, will promote the appearance of more personalized devices, lower-cost solutions, the use of more sustainable feedstock products, and 'greener' alternatives for the end-of-cycle of these products will become available. However, some challenges to the development of these technologies are yet to be overcome.

Some challenges relate to the devices themselves and include the need to assure higher performance of the SCP produced through PE-related technologies, as well as assuring that the power supply and power consumption are optimized for their long-term use. Moreover, since these SCP are intended to be retrieving large amounts of personal data, its management and security will also play a role in the long-term acceptance of these devices by the public. Another concern is the lack of manufacturing standardization and regulation that is inherent to such recent technologies. Skilled workers in the field of PE, capable of implementing such manufacturing standards and creating/obeying regulations are also in demand.

Nonetheless, assisted by R&D in novel materials and technologies, the SCP are set to enhance the functionalities of pre-existing industries, as well as precipitate the debut of new and innovative ones, forever altering society as we currently know it. Through the seamless integration of flexible, lightweight, and conformable electronics this future paradigm no longer looks like the distant reality once pictured in the movies. As for now, value and risk are still being weighted as the SCP reach small ecosystems.

Data availability statement

No new data were created or analysed in this study.

Acknowledgments

This work has been supported by NORTE-06-3559-FSE-000018, integrated in the invitation NORTE-59-2018-41, aiming the Hiring of Highly Qualified Human Resources, co-financed by the Regional Operational Programme of the North 2020, thematic area of Competitiveness and Employment, through the European Social Fund (ESF), and by the scope of projects with references UIDB/05256/2020 and UIDP/05256/2020, financed by FCT—Fundação para a Ciência e Tecnologia, Portugal.

Conflict of interest

The authors declare no conflict of interest.

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