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Influence of design properties of printed electronics on their environmental profile

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Abstract. In the context of an Internet of things (IoT) vision, printed and embedded electronics have gained serious momentum over recent years. Large leaps in innovation promote the applicability of the technology and help reduce device cost significantly. Additionally, printed electronics are often perceived as a green technology with high potential of replacing established subtractive manufacturing methods and act as an enabler in many areas of society. However, their environmental impacts are still rarely investigated thoroughly. Device development for printed electronics typically starts with the definition of functionalities rather than exact knowledge about components and materials, making an integrated early-stage life cycle assessment (LCA) of the devices challenging due to the typically large amount of possible technical solutions for each use case. This contribution fundamentally supports the idea that getting involved with environmental considerations as early as possible in the development is pivotal in avoiding sustainability pitfalls from the start. Consequently, several LCA studies are summarised focusing on three different sustainability scopes: material, production and device, as well as use-phase and end-of-life. The work aims to provide an overview over the sustainability potentials and risks of the production processes of printed electronics from flexible substrates and conductive inks based on micro- and nano-sized particles. Different filler materials for the inks are considered, as their impact heavily influences the overall device impacts. In conclusion, recommendations for further work in the field are derived, summarising potentials of printed electronics, while equally considering remaining challenges. Thus, the conducted work contributes to a better understanding of environmental impacts in the development of printed electronics and helps applying the findings already at the very first development stages.

Keywords: environmental footprint / printed electronics / functional materials / conductive particles and formulations / LCA / design for environment

1 Introduction

Steadily increasing performance of electronic circuits and devices at decreasing cost has led to the integration of electronics in almost every area of human life. Conventional production of electronic devices hereby causes large environmental burdens, particularly due to the subtractive, energy and material-intensive character of transferring functional patterns onto substrates [1,2]. Considering the history of extraordinary growth in the industry of 3 to 5% per year, combined with decreasing device lifetime and low recycling rates [3], unsurprisingly its overall environmental impact has been increasing steadily.

The emerging technology of printed electronics carries the promise to achieve significantly lower costs at higher production speed, while at the same time reducing environmental impacts from production though waste reduction and avoided use of etching chemicals [1,2,4,5]. Generally, the terminology printed electronics refers to additive production processes of electronic components and must not be confused with the production of conventional printed circuit boards (PCBs) from subtractive processes despite similar terminologies [2,6]. More specifically, "printing" in this contribution refers to the additive deposition of functional particles via liquid ink systems onto a variety of substrates in patterns that function as conductor, semi-conductor or insulator in an electronic circuit.

One of the decisive advantages of printed electronics can be a substantial reduction of required manufacturing steps per device in comparison to conventional electronics [1,5,7]. Simultaneously, flexible and stretchable instead of rigid substrates in combination with their ultra-light weight allow for applications that have not been feasible

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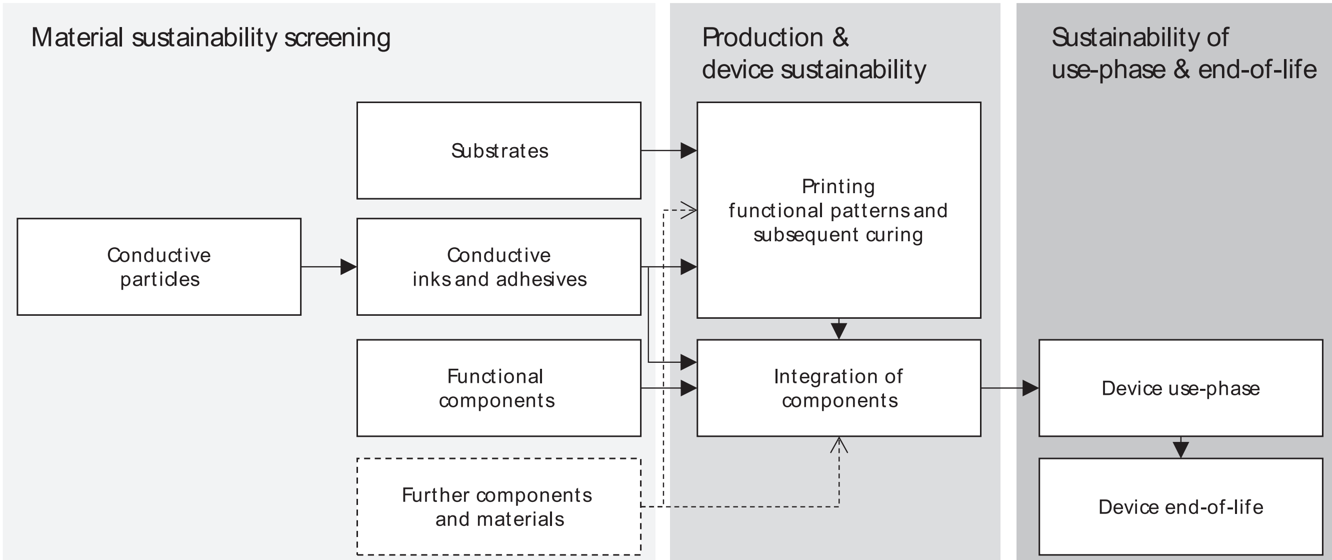

Fig. 1. Overview over the life cycle of printed electronics and the three scopes discussed in this contribution.

before [1,8–10]. These overall trends lead to predictions of extraordinary growth for the printed electronics industry in the coming years, creating a wealth of social and economic impacts [10,11].

Unfortunately, the promising future of printing electronic circuits often conceals what technical challenges are still unresolved [11]. In contrast to conventional manufacturing, environmental impacts for printed electronics also have not been investigated thoroughly through life cycle assessment (LCA) studies to identify sustainability hotspots and the influence of different design properties. Yet, for making sound decisions on business cases, the often-conflicting interests regarding technical performance, economic attractiveness and environmental sustainability of printed electronics need to be weighted and reconciled. While this is by no means a revolutionary approach in the electronics industry [12], and despite an ever-increasing amount of large-scale applications, the sustainability aspect is often underemphasised in current publications on printed electronics.

Particularly, the possibility to predict environmental impacts for devices in early development stages still bares significant challenges for LCA experts as even within the industry a variety of process details seem unresolved [2,10]. Kunnari et al. already presented detailed considerations on the environmental evaluation of printed electronics in 2009 [2], using a conventionally manufactured device as a reference. Also, Wiklund et al. criticise the current practise of adding intelligence into everyday objects through printed electronics without regards to environmental considerations [13]. In their paper, they also refer to several other publications discussing environmental impacts, mostly of singled out applications or devices. To the knowledge of the authors, there has not been any comprehensive and quantitative sustainability overview of materials and processes used for printed electronics.

The publication at hand picks up on detected research gaps using industrial primary data and a combinatory approach to map the range of environmental impacts in which a device is likely to end up. Consequently, this study shines a light onto environmental hotspots and sustainability challenges in printed electronics, and how these can be addressed during device development and design. In general, the evaluation of printed electronics in this paper is divided into five major material segments, the production processes, as well as the use-phase and end-of-life, as illustrated in Figure 1. Detailed life cycle inventories are not disclosed within this publication due to the confidentiality of processes and formulations. However, primary industry data has been crosschecked and verified with published literature to assure high quality throughout all modelling steps.

2 Material sustainability screening

The key to shifting conventional manufacturing towards printed electronics lies in the highly specialised materials used [14]. One decisive material is the substrate to be printed on, while the main focus lies in finding functional electronic materials and respective inks with suitable properties to print them [5]. Conventional printing of information (i.e., text and graphics) is a well-established technology optimised to a high degree, yet conductivity was formerly not a design prerequisite in printing and offers specific issues [5,6]. Most importantly, for all materials, tweaking the properties to the specific use-case and printing technologies is the key to realising novel printed electronic applications [15]. When selecting the materials for one application, several interdependencies need to be considered as not all materials can be combined with each other and their processing plays another crucial role. This section does not discuss possible matching materials but rather aims at providing an overview over materials without any claim to completeness.

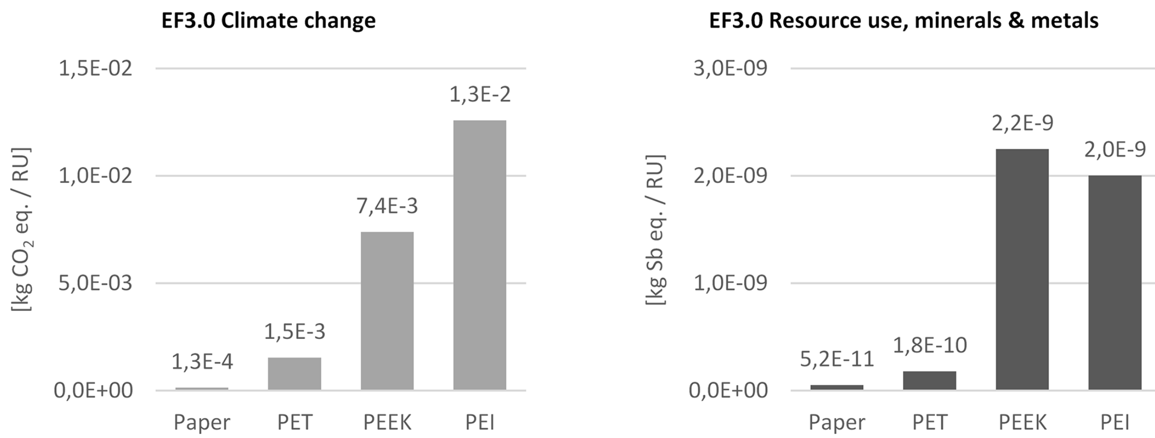

Fig. 2. Contribution to selected environmental impact categories for exemplary substrates (reference unit [RU]: sheet with 25 cm^2 surface area, 125 μm sheet thickness).

2.1 Substrates

It is important to initially consider the material, which is being printed on. Substrates can be either a base material for mechanical stability, a top material for interactive surfaces, or separating interlayers within a printed component [1] and have previously been identified as a key factor for sustainable printed electronics, owing to their significant mass share per device [16]. And while the authors identified conductive inks, and more specifically the contained metal nanoparticles, as a crucially more relevant component [17], they support the efforts in promoting more sustainable substrates.

In theory, any material that has already been printed on in conventional printing processes before, should be considered a potential substrate for printed electronics. Unsurprisingly, beyond glass fibre reinforced epoxy typically used in conventional electronics [1], various substrate materials for printed electronics have been reported. These include paper, plastics, metals and alloys, fibreglass, ultrathin glass sheets, as well as natural rubber, transparent nano-cellulose, and textile fabrics [1,14,18–20]. Beyond that, materials such as gelatine, silk, leather, and even edible caramelised sugar have already been successfully trialled in studies [21]. Currently plastic films, predominantly from polyethylene terephthalate (PET), are most frequently used while also paper offers interesting opportunities [18].

From a performance point, substrates need to be thin and flexible to meet requirements set by the processing [11] as well as subsequently in many applications [20]. The selection is based on a multitude of criteria: processing prerequisites include compatibility with inks and adhesives, the ability to be cut, and temperature resistance, while use-cases influence price and requirements regarding physical and chemical stability, maximum weight, and others [1,19,20]. In addition to those technical and economic properties, environmental impacts should be used as one criterion for decision-making, as different types of materials cause distinct impacts in their production and provision.

A comparison of different common materials for printed electronics is shown in Figure 2 for climate change and resource use, including paper, polyethylene terephthalate (PET), polyether ether ketone (PEEK), and polyetherimide (PEI). The underlying LCA models include the cradle-to-gate production process of substrate without final packaging or transport and are built in GaBi software [22] with the CUP2021.1 database [23].

In the modelling, substrate thickness (125 μm) and measure ($5 \times 5\,cm^2$) are selected for demonstration and do not consider the resulting mechanical properties that might differ between the materials. For all substrates, surface treatment processes like supercalendering, coating and polishing are not included in the presented model. These are expected to moderately increase environmental impacts resulting from production due to additional energy and/or auxiliary material consumption.

The overall environmental impact of the substrate per printed electronic device depends on two aspects: material selection and substrate dimensions. The former is typically based on optical preference and the required stability regarding stress, moisture, temperature, pH and others. The latter is typically determined by the circuit layout and design of the printed device to be produced. Most of the environmental impact stems from the provision of polymer granulate for all three plastic substrates (between 72% and 98% contribution per impact category), and from the processing route for paper substrates.

2.2 Conductive particles

Electronically conducting materials are of central interest, as they bring desired properties (i.e., conductivity) to the device. In principle, any material with sufficient conductivity could be trialled for printed electronics, if it can be formulated into a transport medium. Consequently, a wealth of research has already been concerned with the development of conductive materials [4,5,15]. This specifically includes metals, graphene derivatives and carbon nanotubes (CNTs), as well as carbon-based compounds with a conjugated backbone (i.e., “conductive polymers”, such as PEDOT:PSS) [1,5,8–10,14,15,24–26].

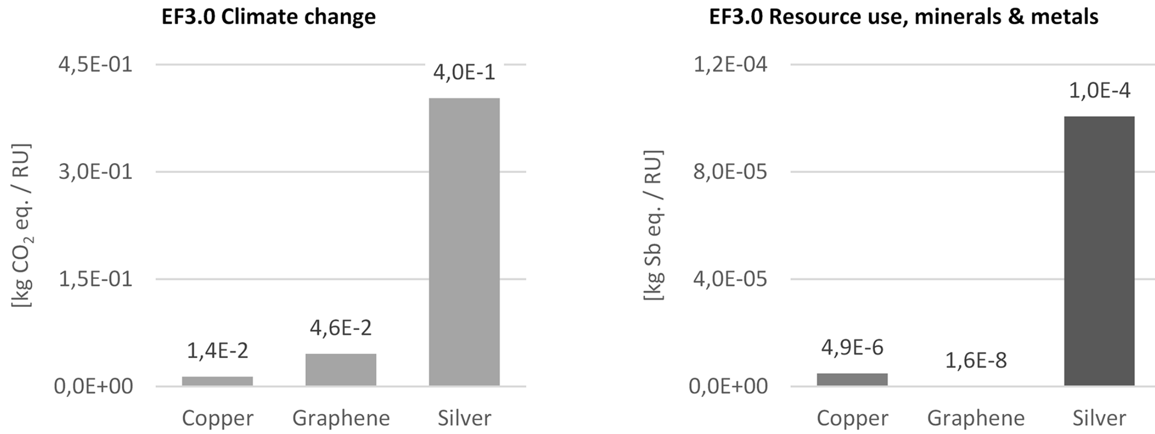

Fig. 3. Contribution to selected environmental impact categories from copper, graphene, and silver nanoparticles (reference unit [RU]: 1 g particles, theoretical dry weight).

In this study, inorganic functional inks are investigated, as manufacturers currently mostly rely on metal materials. Specifically, nano-sized metal materials are preferred for their high conductivity, environmental stability, and easy formulation into required inks [15,20]. The term nanoparticles hereby commonly refers to particles appearing in the nanometre range in at least one external dimension, which can come in a variety of different shapes, such as spheres, flakes, wires, and others. Besides that, also micron-sized particles can be found in printed electronics applications.

Particularly noble metals are sought after for printed electronics, owing to their stability against oxidation, which can hamper conductivity [14]. First and foremost, silver is the most commonly used metal for printed electronics so far [4,10,11,25]. However, a multitude of other materials have been researched. This includes inter alia copper [5,26], gold [5,27], zinc [28], palladium [5,29], aluminium [14], iron [27], as well as different transparent conducting oxides (TCOs) [5]. Some of these materials are rather seldom used owing either to procurement cost or shortcomings in processability and performance [15,25]. The most promising alternatives for silver currently seem to be copper and zinc nanoparticles. They are particularly interesting for their cost saving potentials at comparable conductivities but challenges in material development regarding fast oxidation still need to be overcome [15,25]. Lately, especially for zinc, significant advancements have been reported by Majee et al. [28].

Metal nanoparticles are used far beyond printed electronics and can be produced top-down by breaking down bulk material or bottom-up, using compounds that are reduced in the process [5,25,27]. Hereby, the latter is the preferred approach due to easy control over particle size and shape, which is a decisive factor for device performance [10]. However, Efimov et al. recently argued that top-down production of nano-particles would be more environmentally friendly and less costly [24], unfortunately, without providing quantitative prove for their point in the form of an LCA study. Additionally, biological routes through microorganisms or plant extracts have been reported for silver nanoparticles, where LCA data again is not available [27].

For this contribution, silver and copper nanomaterials obtained in wet-chemical bottom-up processes, as well as graphene from exfoliation are assessed, since they constitute the research focus within the projects so far (Fig. 3). Underlying LCA models are conducted in GaBi [22] and include cradle-to-gate production processes of nanoparticles, including precursors, required chemicals and electricity for processing, as well as packaging in polyethylene bottles. The production processes are regionalised to the location of project partners providing the materials (DK, ES, and GB). LCI data is based on primary sources on lab-scale and the CUP2021.1 database [23] for background modelling. Graphene nanoparticles are modelled as dry powder (exfoliation of graphite and subsequent drying), while the metal nanoparticles are modelled as dispersion of 70 wt% Ag (precipitated from $AgNO_3$) and 65 wt% Cu (from $Cu(OH)_2$) respectively. For easier comparability, the results used in the following comparison are converted to 1 g of dry particle mass. It is important to note, however, that this reference unit (RU) for comparison is rather theoretical, and the performance (i.e., conductivity) differs quite significantly between the materials. Thus, larger amounts might be required for less conductive materials, which coincide to be the more sustainable options. Moreover, the drying process taking place for the graphene makes up more than half of the environmental impacts, somewhat distorting the results.

Largest contributors to resource use for Cu and Ag nanoparticles unsurprisingly are the precursor metal salts (>95% of resource use), while the carbon footprint depends on various factors and metal salts only contribute to roughly one third (27 to 37% of climate change). For the production of graphene nanoparticles, electrical energy consumption on-site is the decisive factor (66% of climate change and 75% of resource use). All revealed data is subject to uncertainty due to single data sources at project

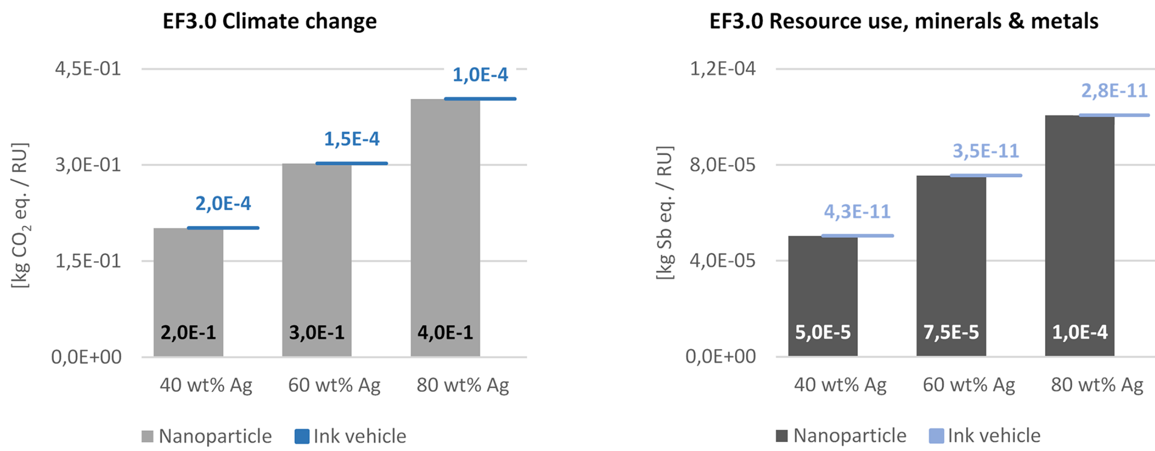

Fig. 4. Comparing selected formulations for water-based, conductive silver inks including additives, and varying filler content (reference unit [RU]: 1 g of ink, wet weight).

partners and the pilot scale of some of the processes. Consequently, further sensitivity studies are highly advised.

Finally, the overall environmental impact of the conductive materials used in functional inks for printed electronic are determined by type and amount of material. While the type of conductive material is typically selected due to price, performance and processing capabilities, the amount is defined by the desired conductivity.

2.3 Conductive inks and adhesives

To transfer conductive particles to the desired locations and form functional patterns, an ink system is normally used [24]. Inks are colloidal solutions of functional materials and binders in an ink vehicle, typically enhanced with various additives such as stabilizing agents, oxidation inhibitors, and surfactants [1,5,11]. This allows tuning ink properties, such as viscosity and filler loading, to the selected printing technology and vice versa, with both depending significantly on the printed electronics application [15]. For printed electronics, conductive inks are predominantly based on nanomaterial from one single metal element [18]. There has been a wealth of applications dispersing them in different types of solvents for printing patterns and interconnections [8], including water, isopropyl alcohol, toluene, xylene, and cyclohexanone [5]. Hereby water is considered more environmentally friendly than organic solvents even though the wetting properties of water-based inks are more challenging [5].

In principle, dry particle deposition after arc or spark ablation from bulk metals and transportation via a continuous gas stream could be an alternative to ink systems [30,31]. Its environmental performance has been analysed by Slotte and Zevenhoven before [31], but needs further attention regarding the comparison between the practice based on wet-chemical processes and the use of carrier gases in combination with arc or spark discharge. This technology therefore remains an interesting field that deserves further research.

A specific way to use conductive particles is in the formulation of interconnector materials [14]. These are vital for system integration within printed electronic devices and a critical weak spot [3], as reliability and stability of interconnections is pivotal to avoid product failure. From the three fundamental types of interconnectors used in electronics manufacturing (soldering, press-fitting and electrically conductive adhesives) only the latter are suitable for printed electronics. Hereby particularly, the low temperature thresholds from the substrates ($\leq$ 80 to 130 °C) are decisive [3]. On top of that, they are considered an environmentally promising alternative to the phased-out lead solders previously used in the electronics industry [14]. Electrically conductive adhesives are inks or pastes composed of a polymeric binder matrix and conductive metal nanoparticles like gold, silver, copper and nickel, analogously to the active media in conductive inks, as well as further additives [3]. While in principal the same considerations apply for adhesives as for inks, the typically much lower amounts required per printed electronic device overcompensate the potentially higher filler loadings by far [17]. This makes inks more relevant in terms of critical hotspots, yet most findings should be transferrable to adhesives analogously.

The sheer variety of possible ink formulations being investigated by other authors or already in the market do not allow for an exhaustive dissemination in the scope of this publication. Therefore, only selected examples are taken into account without claim to represent every existing configuration. Modelling is conducted in GaBi [22] based on primary LCI data from developers, and GaBi CUP2021.1 [23] for background system. Cradle-to-gate modelling of inks includes precursors (nanomaterials as previously described), required chemicals and electricity for processing, as well as packaging in PE bottles. The modelled nanomaterials are simplifying real nanoparticle utilisation, as the particle size and shape partly depends on the desired application. The obtained LCA results for different ink formulations are displayed in Figure 4.

The environmental impacts of conductive inks for printed electronics depend on two characteristics: the overall amount of ink required for the designated patterns as well as the formulation of the inks. Hereby, the environmental impacts of different formulations is almost exclusively defined by the filler loading. For the considered ink the contribution from the Ag nanoparticles is even exceeding 95% for both investigated impact categories. Another factor can be the solvent, since water based inks perform better than inks based on organic solvents, as previously reported [5] and confirmed in own models. This can mainly be attributed to the evaporation of VOCs during printing [2] and the higher environmental impacts that come from organic solvent production. However, for the investigated ink formulation this aspect is negligible due to the high contribution from the filler. The amount of filler is dependent on the application and significantly influences ink viscosity and consequently processing parameters such as adherence, dimensional stability and electrical resistance [14].

2.4 Functional components

The functionality of electronic devices is typically achieved through distinct components offering specific functions such as memory, processing, communication, visual and optical outputs. These can include integrated circuits (ICs/“chips”), LEDs, transistors, resistors, batteries, and many more. Manufacturing those functional components in conventional processes is established and has been optimised to a high degree. Therefore, even for printed electronics, some of these components are often off-the-shelf elements that are manufactured in conventional processes and integrated into the devices [10] as printed devices still form substantial challenges [5].

Owing to their energy intensive production, conventional functional elements can contribute significantly to the overall environmental impacts of a printed electronic device. This has been demonstrated for integrated circuits (ICs) used in RFID transponders with shares of up to 76% in one impact category, depending on the provided IC functionality [17]. The environmental impacts of different conventionally manufactured functional components have been explored in great detail before. For example, conventionally produced silicon ICs have been discussed by Andrae and Andersen [32] and Boyd [33].

On the other hand, significant advancements for fully printed circuits have been made, which carry the promise to replace at least some conventionally manufactured components [34–36]. To this date, however, there still seems to be plenty of functionalities that cannot be fulfilled in a technically adequate and economically attractive way. Moreover, environmental impacts over the entire life cycle of printed components versus conventional components have not been investigated thoroughly and still form a major gap of research.

As of now, most functional components are not (yet) printed and therefore not discussed in the publication at hand. As a first indication of saving potentials, the authors investigated four different RFID transponders including either fully printed, or one of two conventional silicon-based ICs and found a more than 90% lower impact for the fully printed IC [17]. Yet, the study has not been critically reviewed according to the ISO 14040 standard [37], which should be a prerequisite before using the outcomes for further conclusions. Further, it became evident that the trade-off between sustainability and functionality needs to be investigated further before really making comparative statements on their life cycle impacts. Lastly, there is even debates, whether it is desirable at all to shift the production of high-volume, customised devices to fully printing all components [5]. For the foreseeable future, the authors therefore expect both conventional and printed electronics to exist in parallel for different specific applications.

2.5 Further components and materials

There are various other components and materials used in printed electronics. Besides the previously discussed conductive inks, also semi-conductive and dielectric inks play a decisive role in printed electronics [1,11]. So far only one dielectric ink used in printed RFID antennas has been investigated by the authors, which was found to have negligible influence on the overall environmental performance [17]. Further, ferromagnetic shielding layers to reduce interference for wireless communication can be required [38]. These can be printed analogously to the presented considerations for conductive inks building on ferrite nanoparticles. Consequently, the deliberations regarding the environmental impacts of inks can be transferred to this material. Similarly the previously discussed substrates can also be used as conversion layers to protect the printed pattern from mechanical stress and act as a touch surface, or as intermediate layer, e.g., separator membranes in batteries [1]. Again, the environmental considerations stay the same, although the technical application may be fundamentally different. Lastly, encapsulation of entire devices can be used to protect the circuitry, e.g., from moisture and oxygen diffusion [15]. As both, material and geometry, are expected to vary dramatically between different applications, the discussion must be conducted based on particular use-cases.

As all of the above mentioned elements are not necessarily included in every printed electronic device or are simply variations of what has been discussed before, their assessment is not discussed further in the paper at hand. Nonetheless, the materials remain critically relevant for further investigation. First assessments for printed RFID devices indicate the range of possible contributions to the devices’ environmental footprint for these components [17]. Yet, these results should only be seen as a first involvement with the domain and need to be verified for other materials and use-cases, as the devices are little complex in comparison to what is expected to be possible with printed electronics in the future.

3 Production and device sustainability

After reviewing the materials used for printed electronics, it is important to consider the actual production process in which these advanced materials are applied. This includes the printing of functional patterns and subsequent post-processing, as well as the integration of components that may or may not be printed themselves. Lastly, after understanding the drivers within the production process, a cradle-to-gate assessment of device production can be conducted. This, however, still forms challenges due to the large amount of unknown specifications and the variety of possible materials for each end-user application.

3.1 Production processes

Björninen et al. already described the significant influence fabrication methods can exert on functionality and performance of RFID transponders as exemplary electronic devices [39]. A similar influence on performance can be expected to be valid for any type of printed electronics and equally extend into the environmental profile of the device. As printing processes partly define material selection and properties required for processing [1], they consequently significantly influence the environmental profile of devices. Moreover, the production processes themselves also require different amounts of electrical energy and auxiliaries, creating different amounts of waste and emissions.

There is a wealth of different additive printing technologies available such as screen, inkjet, gravure, flexographic, and offset printing [1,5,6,16,40]. One crucial advantage of these technologies is the possibility to produce large area electronics in roll-to-roll (R2R) or sheet to sheet (S2S) processes on flexible and lightweight substrates at low cost [1,11]. Another advantage is the flexibility and degree of individualisation, as small lot sizes can be achieved through digital non-impact technologies such as inkjet [13]. Moreover, printing processes for electronics integrated into additive 3D manufacturing technologies as described inter alia by Maalderink et al. [41] allows for circuits to be literally embedded, unlike with any other production method available to date.

Particularly when compared with conventional subtractive photolithographic manufacturing, additive printed electronics manufacturing offers a significant reduction of process steps, auxiliary demand, and waste generation, a fact often emphasised to present the technology as environmentally superior [1,2,4,5]. However, given the deliberations presented before, it is reasonable to expect significantly different environmental impacts for each processing route and technology within the field of printed electronics. Due to missing knowledge on impacts, the selection of the "right" manufacturing process as of now can be observed to de facto only depend on the device to be produced and readily available technology at each manufacturer.

In principal, the production process for any printed electronic device basically follows the same process steps that can be iterated many times. In a layer-on-layer approach different materials with distinct properties can come to use to finish one product [1]. These crucial steps are:

- *material deposition* [1,6,11]: the actual printing step describes the selective deposition of the functional materials within their respective ink system at exactly the location where they are needed to form functional patterns. This can be performed as a contact-based or non-contact process in one of the above mentioned established printing technologies;
- *post-processing* [1,5,14,15,25]: secondly, a post-treatment process takes place, ideally fully removing the ink vehicle and additives, while also initiating inter-particle necking and fixation of patterns in place to form one continuous phase. This step also serves to achieve desired electrical characteristics and later device functionality through the removal of insulating ink components, thus increasing percolation paths. Post processing can be performed via drying, curing, sintering, chemical transformation, or annealing. For inks based on metal nanoparticles, the process of temporary liquefying solid material through heat, allows forming an interconnected solid layer. Hereby, the effective melting point (T_m) of nano-sized material is drastically lower than for bulk material, making the use of metallic nanoparticles feasible even with some heat-sensitive substrates such as paper and plastics.

If functional components are embedded, the process follows a similar procedure as printing with a step of adhesive deposition and subsequent post-processing. Additionally, the components such as surface mounted devices (SMDs) are positioned in an intermediate step. This can be done, e.g., by a pick-and-place module. However, this process is crucially dependent on alignment and accuracy to ensure functionality of the integrated components. Therefore, it still forms a major challenge within printed electronics. Generally, the combination of different processing technologies still forms challenges due to the required precision to control all parameters at substrate speeds between 5 and 50 m/min [11].

While initial investigations indicate, that the environmental profile of manufacturing processes for printed electronics only has a minor to negligible influence with 10% or less for all investigated EF3.0 impact categories [17], this is only a starting point. The considerations only include R2R printing on one pilot line with water-based silver nanoparticle inks. To produce robust statements on whether the production processes indeed are relevant, further research is required. So far, particularly the consumption of electrical energy was found to determine the environmental profile. However, auxiliaries and waste (particularly the valuable conductive inks) certainly deserve a more thorough investigation. In any case, waste reduction and on-site recycling must be aimed for, wherever possible.

3.2 Combinatory approach for assessing device sustainability

Most frequently, LCA is conducted for one product system at a time with clear system boundaries and product specifications. This focus on a typically small amount of discrete scenarios can be applied straightforwardly for materials and product systems that are close to a market-ready composition or even have a bill of materials readily available already. As the applications for printed electronics are still in a very dynamic stage of development, it seems unwise to wait for final product specifications before investigating the device sustainability. Much rather, printed electronics offer the rare chance to influence material and device development through an early stage complementary environmental assessment, making it likely to avoid a significant amount of impacts at the design stage [2].

For assessing recycling potentials and challenges, the reference unit of "one generic device", which includes a multitude of properties typically found in printed electronics, has been suggested before [42]. However, this approach rather allows for a perspective on printed electronics as a whole and seems less suitable for assessing hotspots resulting from design properties on device level. In a different context, a more promising approach has been suggested by Betten et al. for personalised use-phase assessments in LCA [43]. They argue that, while combinatorics is not a complicated approach, it is currently not exploited enough in LCA, even though there are hardly computational restraints in terms of scenario numbers.

Combinatorics might help solve a dilemma that is present for making sustainability-based decisions in development and design of printed electronics. As one would expect, developers and manufacturers typically only have a vague idea of the environmental impacts their products can have, and what design properties exert which influence. Often sustainability performance is evaluated in comparison to conventional manufacturing and based on life cycle inventory (LCI) metrics such as amount of generated waste, amount of used auxiliaries (e.g., etching chemicals), and so on, typically expressed per device. While these metrics do translate into LCA impact categories such as climate change and resource use, it is close to impossible for developers and manufacturers to accurately quantify these without LCA expert knowledge. On the other hand, LCA professionals may be able to quantify impacts with a high degree of certainty, yet they typically lack the deep understanding of the product system, as there is a multitude of disciplines involved and interdependencies between almost all design parameters exist.

On the way towards comprehensible environmental assessments of printed electronics, a visual approach to illustrate the influence of design properties in early development stages and LCA-inherent uncertainties as suggested by Pfeuffer et al. [44] seems very promising. Further, as use-cases play a paramount role in defining functionalities, and functionalities define materials and design properties, a user-centric approach seems indispensable. In this field, Briem et al. have established first approaches how to address the topic within the methodological LCA framework [45,46]. While their focus is on use-phases with large influence on the overall product sustainability, it seems plausible to transfer the general concept of highly individualised assessments to printed electronics, where the use-phase is expected to be responsible for the least impacts, particularly when considering single-use PE devices.

The vision is to already use first concepts of a system design to determine the order of magnitude for environmental impacts. At this stage, the approach also offers the possibility to determine hotspots in an explorative manner. As the knowledge on device specifications increases through ongoing development work, the environmental impacts can be narrowed down further. Currently, this combinatory individualistic assessment is developed further by the authors to allow for quick and reliable insights into sustainability of printed electronic devices. First considerations for such an early-stage assessment were presented by the authors [47].

Similarly to what Betz et al. already proposed more than 20 years back for the electronics industry in general, a need for homogenous data collected in cooperation between research and industry is a prerequisite to proceed with sound assessments of printed electronics [12].

4 Sustainability of use-phase and end-of-life

Finally, it is worth to mention some aspects on the life cycle phases following production. Owing to the functionality requirements for the use-phase, materials and processing technologies are selected, which then crucially influence what happens when the operating life of a device comes to an end. Consequently, both phases need to be equally considered to avoid sustainability pitfalls early on.

During the *use-phase* of printed electronics, there is no expected direct environmental impacts, if the printed devices do not consume electrical energy, and peripheral power supply (e.g., via radio waves) is disregarded [17]. This is certainly a simplification that needs to be reconsidered, in particular for devices with extended lifetime as this would make power supply increasingly relevant. In this case, a change of the LCA system boundaries is required to include out-of-device power consumption for a holistic picture. Currently, the authors are not aware of any study that has used such a wide scope for the environmental assessment of printed electronics.

Apart from this important consideration, the implementation of PE into other products or product systems (i.e., into the "things" in an internet of things) could have significant benefits regarding the performance of the equipped products [48]. RFID technology, for instance, can have possible impacts on various levels of the equipped products' life cycle [17]. These could occur during production, in the use-phase, or influencing the ability to be treated at the end-of-life, as well as life cycle costs. Indirect impacts from rolling out electronics into society must not be ignored, as postulated by Moreau et al., yet tools need to be available [48]. This also includes negative impacts, which seem just as plausible for some applications.

The authors already proposed some general advice for selecting suitable applications of printed electronics [17], arguing that the decision to implement printed electronics should always be based on the potential to reduce energy or material consumption of the equipped product system, or overall transportation efforts. In other words, if no environmental benefits seem feasible through the implementation of printed electronics, there need to be strong arguments to proceed anyways. Particularly use-cases where equal functionality can be achieved through alternative technologies, e.g., printed QR codes instead of passive RFID transponders, the environmental performance should be the decisive argument.

Above all, the device *end-of-life* for printed electronics is crucial due to valuable materials contained. While some substrates can be recycled, and in the case of paper even composted [16], metals such as silver, copper, or others form a significant challenge. Generally, for all electronic devices and components, these resources play a paramount role in achieving the desired functionality [49]. Even though their material content per device is relatively small (e.g., 25 mg Ag per RFID antenna [17]), a mass application in an "internet of things" can lead to serious effects on raw material demand and in consequence threatening supply security [50,51]. Even more problematic than the sheer amount of valuables for all devices combined is the unclear path they will take once their operating life ends. For RFID transponders, the most likely end-of-life scenario is expected to be a collection as part of the mixed municipal waste flows, disregarding the applied production technology [51]. This also seems a likely path for other types of printed electronics.

Particularly the short operating life of devices needs to be seen as a crucial issue from an eco-design perspective [2]. And printing with metal nano-particles strongly contradicts current considerations, e.g., by Dominish et al., on how to achieve increasing material efficiency in electronic consumer goods [49]. And although biodegradability and biocompatibility have already been declared requirements to achieve sustainable electronics in the future [21], this paradigm is diametrically opposed to the widespread use of metal nano-particles in single-use devices practised today.

Whenever metals are used within (printed) electronic devices, their concentration is characteristically low due to the desire to cut costs per device. These low contents, however, often hamper cost-effective recovery and ultimately lead to the loss of material from recycling [52]. This effect of material dissipation, sometimes also described as a "dilution problem", paired with extensive numbers of devices forms one of the most relevant, if not the most relevant sustainability issue for printed electronics. A first initial screening showed that material recycling would drastically cut the environmental impacts in comparison to landfilling or thermal treatment [17]. As separate collection of printed electronic devices for recycling seems unrealistic to roll-out, material alternatives will become decisive for an environmentally responsible future of printed electronics. This has never been more relevant than for 3D printed objects with fully embedded circuits, where it seems impossible to visually detect if electronic components are integrated into the object. Alarmingly, to date there is no consensus on how to include dissipation into LCA studies [52] and only very recent efforts to quantify dissipative losses via material flow analysis (MFA) [53], rendering fact-based decision making challenging for printed electronics developers as of now.

For both, the indirect impacts during the device use-phase and the continuous material dissipation at the device end-of-life further research is urgently required. It is to be expected that early critical discussion on the topic can avoid a major sustainability pitfall of the still maturing technology. The authors suggest to hereby set the focus narrow and application-specific for the use-phase, and more general and material-focussed for the end-of-life phase. Particularly, alternative and more sustainable materials should be a central focus of research. Notably, even from industry associations, concerns regarding future environmental regulations, including a ban of some materials for printed electronics has already been articulated [42].

5 Discussion

The presented results clearly show environmentally preferable solutions for suitable materials for printed electronics. It is important to consider that the assessment was based on one production route and location for each material (i.e., substrate, nanoparticle, ink formulation) and is expected to vary when other production routes are considered. Moreover, there are always technical and functional aspects to consider when deciding on the material to use. For example paper may have the lowest CO_2-footprint of the investigated substrates, yet it can hardly withstand moisture, let alone wetness. And if elevated temperatures are required, not even PET might be suitable. Similarly, graphene and copper nanoparticles might show lower resource use, while more of the same material is needed to achieve similar conductivities as with silver, if equal functionality is even possible. Likewise, fully printed electronic circuits seem to avoid significant environmental impacts during production. On the other hand, the functionality is often still limited to more basic functions. In consequence, it seems unavoidable to select the most environmentally friendly material only after having excluded solutions that create major technical issues for the desired application.

In the presented work, the environmental impacts and potentials of printing electronics were addressed for all three life cycle stages. Most of the investigated literature sources strictly focus on the production phase when concluding that printed electronics are more sustainable than conventional manufacturing. A detailed understanding of the production phase of electronic devices is important, as printed electronics perform significantly better than conventionally manufactured ones. However, this contribution also shows that use-phase and particularly the end-of life are just as important. Unfortunately, this aspect is often underrepresented.

Little surprising, one of the most promising approaches to limit environmental impacts of printed electronic devices seems to be the reduction of material consumption per device. Motivated by an equally attractive cost reduction, ink consumption reduction through adapted pattern designs have been trialled for some time now [54,55]. Both papers found a significant reduction potential of deposited ink around 50% that came with only minor functionality decreases and could even be performed by experts in conventional printing with no significant knowledge about electronic devices. Given that environmental impacts could be reduced equally for specific materials, this concept seems to hold tremendous saving potentials per device and needs to be investigated further. That being said, it is critical that this reduction of environmental impacts only relates to the production phase, and lower content of valuables such as silver bare enormous challenges when it comes to device end-of-life.

Lastly more transparency and the consideration of historical trends of technical systems is required for more fruitful discussion on device sustainability, a point previously stated in the context of consumer electronics by Andrae and Andersen [56]. In this spirit, the authors encourage researchers in the field of materials, formulations, printing technologies and electronic devices to publish their findings more transparently to facilitate comparability of materials, formulations and devices. This could help overcome the limited knowledge of single players described by Garcia Bardon, which she attributes to confidentiality and legal challenges. She also claims that this together with complex fabrication and the high number of stakeholders is the reason, why up-to-date LCA databases on electronics are very limited today, as reported by Moreau et al. [48].

6 Conclusion

In conclusion, the authors want to stress that many factors contribute to the environmental profile of printed electronic devices. Therefore, an early screening of environmental implications is vital in guiding product development towards sustainability in the sense of product stewardship. A potential approach for an early-stage sustainability screening has been proposed by the authors in a contribution to SAM 2021 Conference [47].

The work at hand opened up several fields of discussion and identified research gaps for proceeding in the sustainability assessment of printed electronics. Generally, key to sustainable printed electronics is selecting the least environmentally harmful materials and production routes, while still fulfilling the predefined device functionality. In other words, the best technological solution needs to be matched with the best environmental performance and no general answer as to "how green printed electronics are" can be given. Thus, sustainability needs to be verified for each single application of printed electronics. Hereby, three fundamental statements for each of the life cycle phases can be derived from the contribution:

– *production*: an increase in sustainability performance caused by the novel additive manufacturing paradigm is expected. Yet, individual functionality requirements for each use-case demand customised designs to limit environmental impacts and avoid over-engineering. The biggest hotspots currently lie in conductive inks based on metal nanoparticles, as well as off-the-shelf functional components;
– *use-phase*: potential positive influence on equipped product systems during the printed electronics' lifetime is pivotal for their overall environmental performance. These benefits should always exist to justify environmental burdens from the device end-of-life. Unfortunately, especially low-cost, single-use devices produced in large volumes remain a key sustainability threat in that sense;
– *end-of-life*: significant sustainability risks can be expected at the end of the device lifetime associated to the dissipation of valuable metals. One key to tackle this issue is moving away from silver-based inks towards environmentally less harmful solutions.

Moving forward, the authors highly encourage the investigation of additional conductive materials (metal and organic), as well as production processes for printed electronics to gain a more holistic understanding and provide developers with knowledge on occurring environmental implications. As conductive inks have been identified to be one key source for environmental impacts, it is pivotal to continue investigating the sustainability of potential fillers, additives, and ink vehicles, as well as their contribution to the overall environmental profile of the ink. One of the most important aspects to these evaluations is a transparent and open data exchange with researchers and manufacturers from the realm of printed electronics. Lastly, the investigation of further use-cases should be a focus in upcoming research to ensure that printed electronics can contribute to some sort of environmental purpose and fulfil their promise towards more sustainable electronics.

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