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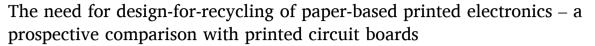
Contents lists available at ScienceDirect

Resources, Conservation & Recycling

journal homepage: www.elsevier.com/locate/resconrec



Full length article



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ARTICLE INFO

Keywords:
Printed electronics
Sustainable electronics
Recycling
E-waste
WEEE
Carbon footprint

ABSTRACT

The present study compares conventional printed circuit boards (having glass-fibre and epoxy substrates and etched copper circuits) with paper-based printed electronics (offering flexible, bio-based, and biodegradable substrates with circuit design printed using silver-based inks) and assesses the relevance of e-waste recycling to the latter's sustainability. Therefore, a comparative life cycle assessment between these two options has been undertaken and the global warming impacts were calculated.

The impact assessment results underscore that printed electronics offer a consistent sustainability advantage over printed circuit boards only through recycling of silver in the former at the end-of-life. Hence, design-for-recycling and recycling as e-waste are crucial to the sustainability of the current generation of printed electronics. Other foreseen waste treatment options for paper-based printed electronics, such as composting, and paper recycling, are likely to limit the sustainability advantage of printed electronics to circuits with small conductive areas.

List of Abbreviations

CNC Cellulose Nanocrystals

EoL End-of-Life

LCA Life Cycle Assessment LCI Life Cycle Inventory PCB Printed Circuit Board

WEEE Waste of Electrical and Electronic Equipment

FR4 Flame Retardant 4

Ag Silver
Cu Copper
Sn Tin
Pb Lead

1. Introduction

Electronics form the backbone of modern-day society and their integration into different aspects of human life is only expected to increase with further advancements in the Internet-of-Things (IoT) (Roselli et al., 2015). The physical world of the future is anticipated to be more interconnected with the digital world (Wiklund et al., 2021) thanks to "smart" everyday objects, such as textiles, packaging, etc.,

featuring enhanced functionalities through the integration of electronics (Glogic et al., 2021). Such smart objects will make electronics ubiquitous, will boost the demand for circuits providing electronic functionality, and also create a lot more waste of electrical and electronic equipment (WEEE) (Hakola et al., 2021). Conventional printed circuit boards (PCBs) may not sustainably fulfil this boosted demand in electronics, considering that PCBs are energy-intensive to manufacture, use hazardous substances, and have limited recyclability (Zeng and Li, 2016). Many of these limitations of conventional PCBs are overcome in novel 'printed electronics' that incorporate eco-design principles (J. Li et al., 2015) and minimize energy utilization during the manufacturing phase (Kunnari et al., 2009). Furthermore, printed electronics, with a multitude of applications such as displays, sensors, electronics, and radio tags, may be manufactured at lower costs and with high productivity using continuous production processes (Bonnassieux et al., 2021; Glogic et al., 2021). Considering these advantages, printed electronics are positioned as sustainable alternatives to conventional PCBs in smart objects, i.e. low voltage and low-frequency applications expected to be manufactured, used, and disposed of in large volumes (Liu et al., 2014; Wiklund et al., 2021).

The distinction between the manufacturing of PCBs and printed electronics is depicted in Fig. 1. As shown in Fig. 1(a), this study

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considers conventional flame retardant 4 (FR4)-based PCBs that utilize a glass-fibre and epoxy resin composite as the substrate. The entire surface of the FR4 substrate is coated or laminated with a sheet of conductive copper (Cu) under high temperature and pressure. Additionally, a chemically resistant masque is deposited on the conductive surface according to the circuit design. Finally, the excess Cu is etched away to reveal the conductive circuit design by submerging the FR4 laminated with Cu in a chemical bath (Esfandyari et al., 2015; Nassajfar et al., 2021; Patel, 2018). Conceptually, the manufacturing of PCBs involves the removal of excess Cu through etching or lithography and is therefore classified as a subtractive manufacturing process (Kunnari et al., 2009).

In contrast to the subtractive manufacturing of PCBs, the manufacturing of printed electronics is achieved through additive manufacturing as it involves only utilizing the requisite amounts of material for the conductive tracks. Additive manufacturing avoids the use and removal (wastage) of excess material, minimizes the use of chemicals by eliminating the etching process (Kunnari et al., 2009), consequently is resource-efficient, and has a lower fabrication cost (Wiklund et al., 2021). In printed electronics, traditional printing technologies are used to directly print intricate circuit designs on a variety of substrates using conductive inks. Thus, continuous manufacturing of printed electronics in large volumes, with high productivity, resource efficiency, and low costs can theoretically be achieved through a setup similar to an industrial printing system applied, for example, to publish books (Glogic et al., 2021). Printed electronics may utilize a variety of substrates depending on the required application, however, this study focuses on the additive manufacturing of a particular paper-based printed electronic system used in the project "GREENSENSE" (LEITAT, 2018) as shown in Fig. 1(b). The GREENSENSE biosensing platform is based on printed electronics and designed for tackling the issue of drug abuse in society through semi-quantitative drug detection in bio-fluids. The functional circuitry of GREENSENSE utilizes paper coated with cellulose nanocrystals (CNC) as a substrate on which the circuit design is printed directly using nanosilver conductive inks (Ag-ink) as illustrated in Fig. 1(b).

Paper is increasingly garnering attention in the printed electronics sector because it can allow for flexible, foldable, light-weight, degradable, and cheap electronics operating at low voltages and frequencies (Liu et al., 2014; Wu et al., 2014). Nevertheless printing the circuit design with the Ag-ink directly on paper is unsuitable due to the latter's high surface roughness, porosity, and wettability; the resulting circuit would have poor performance and short life (Wiklund et al., 2021; Wu et al., 2014). Hence, it is suitable to coat or laminate the surface of the

paper (in this case with CNC) to improve its properties and prepare it for the printing of the circuit design with the Ag-ink.

With the growth in the field of printed electronics, the question of replacing conventional PCBs with novel printed electronics arises. Given that the fabrication of FR4 is energy-intensive, utilizes fossil materials such as epoxy-resin, has limited recyclability, and that etching the circuit design requires substantial chemical and energy input (Nassajfar et al., 2021; Premur et al., 2016), printed electronics seem to trump PCBs in the context of sustainability. Printed electronics offer the possibility of electronics that are resource-efficient to manufacture and have non-fossil, renewable, biodegradable, and biocompatible substrates that are free from toxic flame retardants (Irimia-Vladu, 2014; W. Li et al., 2020; Liu et al., 2014).

Apart from the perceived sustainability benefits, comparing the functionality and performance of PCBs with printed electronics is relevant. As shown in Fig. 1, the printed electronic circuit has a higher resistivity than its PCB counterpart, which indicates that direct replacement of the latter by the former in all applications may not be suitable. Additionally, complex multilayer circuit designs cannot be realized in printed electronics as presently the technology is still in its infancy. The reliability of printed electronics circuits is also not at par with conventional PCBs as faster degradation of the conductive tracks in the former implies a short lifespan or operational life. Consequently, the short lifespan will not elicit regular repairs and maintenance (Keskinen, 2012; Kunnari et al., 2009). However, even when necessary, repairs and replacements would be difficult because electronics components (microchips, batteries, displays, etc.) are mounted on the printed electronic substrate using a conductive adhesive (Ag-based); thus, a component (that is essentially glued-on) would have to be forcefully removed for replacement and this could damage the circuit.

Conventional PCBs have the components soldered on the circuit: a hot metal liquid alloy is applied that solidifies when cooled down and fixes the electronic components on the surface of the PCB. By applying heat, it is possible to re-melt the solder and easily unmount and replace any component. Soldering on components on printed electronics is rarely feasible because the heated solder would damage the biological substrate and lamination: in the case of GREENSENSE, the paper, and the CNC would likely burn. Therefore, conductive adhesives are suitable for most printed electronic applications as they can mount electronic components within the ideal temperature range for the substrate.

The shortcomings of printed electronics such as low reliability, short lifespan, lower performance, complexity limitations, limited reparability, and maintenance also contradict the principles of circular

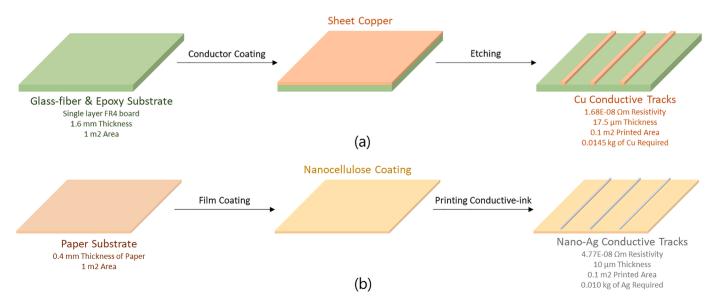


Fig. 1. Conceptual depiction of (a) subtractive manufacturing of PCBs and (b) additive manufacturing of printed electronics.

economy for electronics (O'Connor et al., 2016). Nevertheless, the printed electronics technology is still at a nascent stage and is expected to improve with time (Glogic et al., 2021). Moreover, there are certain low voltage and low-frequency applications that are produced in large volumes but have short lifespans and require simplistic single-layer circuits and in such applications, PCBs will simply not be sustainable (Keskinen, 2012; Kunnari et al., 2009; Liu et al., 2014). In these specific applications, fire risks would be negligible and the 'flame retardant free' printed electronics may be viewed as technically viable, safe, and sustainable alternatives to conventional PCBs. Therefore, a comparative life cycle assessment for the two technologies is warranted to validate the possible sustainability advantage of printed electronics.

Available literature already covers the general LCA of printed electronics (Välimäki et al., 2020), and the LCA of flexible electronics that are incinerated at their End-of-Life (EoL) (Wan et al., 2015, 2017). Moreover, comparative LCAs between printed or flexible electronics and (traditional) PCBs have also been published, all of which found the former to pose lower environmental impacts while briefly considering as EoL options: incineration and landfilling (Kanth et al., 2012); recycling of recyclable components (Hakola et al., 2021); classical WEEE recycling (Nassajfar et al., 2021); and paper recycling if paper is used as substrate (Glogic et al., 2021; Liu et al., 2014). The present study not only compares printed electronics with PBCs but also considers the EoL in detail to explore the relevance and need for recycling the former.

A widely acknowledged concern regarding printed electronics is the massive quantities of e-waste produced because the suitable applications will primarily be low-cost and single-use products that are produced in large volumes, do not require high reliability, and have short lifespans (Keskinen, 2012; Kunnari et al., 2009). Such products would also be plagued with the rebound effect from a cheap technology and be difficult to effectively collect at the EoL for waste management (Galvin, 2015; Hilty et al., 2006). Hence, there has been a focus on the development of compostable electronics by utilizing biodegradable and

biocompatible materials (such as paper) so that accidental mismanagement at the EoL and the consequent leakage to the environment does not create massive piles of persistent and toxic e-waste (Hakola et al., 2021; Shittu et al., 2021; Wiklund et al., 2021).

As a consequence, many bio-based printed electronic systems may be considered as either compostable waste, paper waste, or e-waste at the EoL. But which of these approaches is the best from an environmental and sustainability perspective? Composting of large volumes of printed electronics at EoL poses a resource efficiency concern as the precious silver (Ag) contained in the conductive tracks and the adhesive is lost (Kunnari et al., 2009). On the other hand, the paper recycling system can handle printed electronic waste and recycle the paper substrate (Hakola et al., 2021), but again Ag will probably be lost to the deinking waste stream. In contrast, WEEE recycling will only recover Ag and other metals, but not recover paper. There are uncertainties about which amongst these three possibilities is the best EoL option, and this study aims to resolve this uncertainty. Identifying the most sensible EoL option for an early stage of the technology like printed electronics can guide the product developers to incorporate designs with sustainability and EoL in mind. The field of printed electronics is expected to grow along with the corresponding waste streams (Wiklund et al., 2021), so it is better to deduce the appropriateness of recycling already when the technology is at an early stage to avoid the possible waste issues when the technology reaches maturity.

2. Materials and methods

2.1. Life cycle system

The goal and scope of the comparative LCA are illustrated in the life cycle system in Fig. 2. This study performs a cradle-to-grave (Schmidt and Pizzol, 2014; Zheng et al., 2018) LCA to compare the environmental impacts over the life cycle of 1 m2 of PCB and paper-based printed

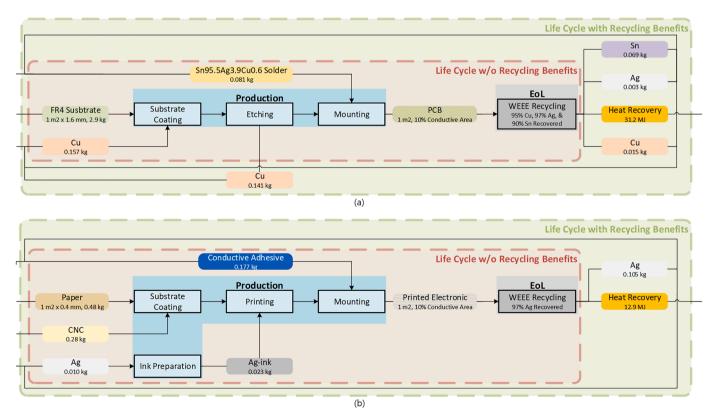


Fig. 2. Life cycle system from cradle to grave with system expansion consisting of recycled materials for 1 m2 (a) Printed Circuit Boards (PCBs) and (b) Printed Electronics with 10% conductive area; the recovery rates of 95%, 97%, and 90% from WEEE recycling for Cu, Ag and Sn respectively, are taken from literature (Huisman, 2003).

electronics with 10% (0.1 m2) of the conductive area. The technical specifications considered for the PCB and the printed electronics have been listed in Fig. 1, and it highlights the lower resistivity of the bulk-Cu tracks in PCBs in comparison to the nano-Ag tracks in printed electronics. Therefore, it is not possible to replace PCBs with printed electronics in all applications because PCBs with a lower circuit resistivity have consequently a higher functionality. Nevertheless, there are some low voltage applications (Liu et al., 2014) in which the higher circuit resistivity of the printed electronics will be inconsequential during use; in such applications, PCBs may be replaced by printed electronics as the higher functionality of the former may be "overkill", i.e. not translate into any real-life advantages in the application phase. Hence, a general comparison between PCBs and printed electronics is justifiable, where the life cycles of only the following essential components differing between PCBs and printed electronics are considered:

- Substrate materials: FR4 in the case of PCBs and CNC-coated paper considered for printed electronics
- Conductive materials: bulk Cu is used in the conductive tracks and a Tin-based (Sn-based) and Lead-free (Pb-free) solder is used for mounting components on PCBs (Adie et al., 2017); whereas for printed electronics, the nano-Ag ink is used for printing the circuit design on the CNC coated paper substrate and an Ag-based conductive adhesive is used for mounting of the electronic components

Unlike the substrate and conductive materials, the electronic components that are mounted on the circuits vary drastically depending on the specific application of the electronics. Nevertheless, these mounted components and their environmental impacts are expected to be identical between PCBs and printed electronics intended (to be replaceable with one another) for the same use. Hence, the different kinds of components and their impacts have been excluded from the scope of this assessment since they cannot assist in distinguishing and further comparing PCBs and printed electronics.

Fig. 2 shows that the life cycle system includes only production and EoL. The specific electronic application (i.e. the use-phase) and the related environmental impacts will be similar between the two options; thus, the use-phase, just like the mounted electronic components, has been excluded from this comparative assessment. The production of PCBs (shown in Fig. 2(a)) starts with coating the FR4 with sheet copper. Following this, the etching process removes the excess Cu to reveal the circuit design of the PCB. The final step of the production process is applying the solder to enable mounting of components, and 1 m2 of PCB with 10% conductive area is ready for use. For the final phase of the life cycle of the PCB (i.e. EoL), proper disposal and WEEE recycling have been considered: this is an "optimistic" EoL treatment for PCBs because it is not always possible in reality and mismanagement of e-waste is common (Galvin, 2015; Hakola et al., 2021; Hilty et al., 2006). During WEEE recycling in metal smelters, the fates of the substrate and the conductive materials differ: the epoxy content of the FR4 is combusted and generates heat; the metallic conductive materials in the conductive tracks and the solder are recovered as recycled products based on the respective recycling rates of metals (Huisman, 2003).

A similar cradle-to-grave life cycle system for paper-based printed electronics covering only production and EoL is shown in Fig. 2(b). The production of the printed electronic involves the coating of a paper substrate with CNC to facilitate the printing of the circuit design with the Ag-ink. Finally, the Ag-based conductive adhesive is applied for mounting the components on the paper circuit. The WEEE recycling considered at the EoL of printed electronics is identical to the one treating conventional PCB-based e-waste and it allows for recycling the Ag, while the paper and CNC are combusted and release energy.

The cradle-to-grave life cycle system described above would only capture the environmental impacts from the production process and EoL treatment. However, the metals are recycled at the EoL and may be considered circular, because the recycled metals may replace some

primary products in the market and offer an environmental advantage. This advantage may be captured by applying the system expansion approach (Nguyen and Hermansen, 2012) to the existing cradle-to-grave model to emulate a cradle-to-cradle life cycle. Thus two possible scenarios are considered in the comparative LCA of PCBs and printed electronics:

- Life cycle without recycling benefits: only the impacts of the production processes and EoL treatment (combustion of non-metallic feedstock in WEEE smelters) are accounted for in this scenario; it realistically captures the impacts over the life cycle for comparison and can serve as a proxy for incineration at EoL scenario (without metal recovery at EoL)
- Life cycle with recycling benefits: assumes that the quantity of metals recycled at the EoL (or also from the etching of PCBs) replaces an identical quantity of primary metals used in the production process; as a result, the recycled metals theoretically offer environmental benefits and assist in reducing the environmental impacts in life cycle system; this scenario may be realized by ensuring the recycling of materials at the EoL and using recycled materials in the production process

Both the above scenarios are considered in this assessment to determine the relevance of WEEE recycling in the lifecycle of both PCBs and printed electronics. Thus, the 'life cycle without recycling benefits' serves as a control and reference for this assessment, against which the benefits of WEEE recycling are gauged.

2.2. Modelling life cycle inventories and impact assessment

The life cycle systems for PCBs and printed electronics in Fig. 2 are modelled using version 3.7.1 of the ecoinvent database (Wernet et al., 2016). To model the production of PCBs, the PCB dataset (Hischier et al., 2007) with the lowest environmental impact was selected. Since this dataset was for a multi-layer PCB including the conductor (Nassajfar et al., 2021), it was adapted to separately model the impacts of the FR4 substrate by removing the Cu input and considering only 80% of impacts from the remaining exchanges. In addition to the production of FR4, the amount of sheet copper required to laminate the FR4 in low-performance PCBs was estimated to be 0.157 kg as shown in Fig. 2 (a), by assuming the (lowest possible) height of the Cu layer as 17.5 μm (Sunstone Circuits, 2022). No dataset exists in ecoinvent to model the etching process, therefore the electrolytic refining activity of Cu was taken as a reasonable proxy to model an electrolytic etching process that removed 90% of the Cu from the PCB surface (Saha et al., 2020). Ecoinvent (Hischier et al., 2007) reports that 0.081 kg of solder is required to mount components on 1 m2 of PCB, so 0.081 kg of Sn95.5Ag3.9Cu0.6 solder paste was selected for this study as that dataset had the lowest environmental impact. The intention behind selecting datasets (for PCB and solder) with the lowest impacts is twofold: (i) to favour PCBs in the comparative LCA, and (ii) to realistically model the low-performance PCBs that can be replaced by printed electronics. The intention behind favouring the PCBs in this assessment is to avoid the influence of any confirmation bias, such as those relating to the perceived sustainability of printed electronics, on the final results.

To model the paper-based printed electronics, data was collected from the organizations involved in the production of GREENSENSE. Process information regarding the material inputs, energy requirements, waste generation, and other emissions was gathered through surveys. Datasets were created using the background ecoinvent data for the various production processes as depicted in Fig. 2(b) – i.e. manufacturing of the CNC, coating of paper with CNC, manufacturing of the Ag-ink, printing the circuit using the Ag-ink on the CNC-coated paper, and application of conductive adhesive. The quantities of paper, CNC, Ag-ink, and conductive adhesive required for a single printed electronic device were extrapolated linearly to quantities listed

in Fig. 2(b) for 1 m2 of a paper-based printed electronic with 10% conductive area. It is important to mention that two configurations of the device have been developed by the GREENSENSE partners; therefore, again to favour PCBs in this assessment, the printed electronic configuration utilizing the higher quantity of materials was selected here for the comparison. Another relevant aspect is that the manufacturing processes modelled for printed electronics were at a labor pilot-scale and thus the corresponding data represented unoptimized processes. This again gives more leverage to PCBs because the impacts from the optimized industrial-scale production (Piccinno et al., 2016) of PCBs are compared to the lab- or pilot-scale production of printed electronics.

For modelling the EoL, standardized WEEE treatment datasets were taken from ecoinvent. These datasets model the recycling and recovery of metals in smelters that manage WEEE waste (Classen et al., 2009). The WEEE treatment datasets require the quantities of metals obtained from the recycling process as input. To estimate these quantities the recycling rates for various metals that are embedded in the WEEE recycling datasets (Classen et al., 2009; Huisman, 2003) were applied and are depicted in Fig. 2. For assessing the recycling benefits in the 'Life cycle with recycling benefits' scenario, the 'market' datasets for the recycled metals and energy recovered were selected to account for the replacement of the primary materials in the market and the generation of heat in the smelter respectively.

All the LCIs were modelled within the Activity-Browser (Steubing et al., 2020) framework of Brightway2 (Mutel, 2017). Finally, the total climate change impacts as per the ILCD's 2018 method (Fazio et al., 2018) were assessed.

2.3. Sensitivity analysis

Since the amount of Ag-ink used would vary with the coverage of the conductive area (Nassajfar et al., 2021), a sensitivity analysis was undertaken to understand the bearing of the conductive area on the comparison of PCBs with printed electronics. For this sensitivity analysis, the 10% conductive area on the circuit board (used in the comparative LCA) was taken as the baseline. Furthermore, the impacts from printed electronics and PCBs were calculated and compared for the following shares or coverage of conductive areas: 25%, 50%, and 75%.

The mass of the Cu etched away in a PCB and Ag-ink required in the printed electronic for the different shares of the conductive areas were linearly extrapolated and have been represented in Table 1. The aim of the sensitivity analysis is also to compare additive and subtractive manufacturing from a sustainability perspective: Table 1 shows that the quantity of Ag-ink required for the additive manufacturing of printed electronics increases with the increased conductive area; in contrast, the mass of Cu to be etched away decreases with the increase in conductive area for the subtractive manufacturing of PCBs. Throughout the sensitivity assessment, the quantity of solder and the conductive adhesives applied for component mounting were kept constant at the baseline levels depicted in Fig. 2. Therefore, the sensitivity assessment is only performed for the conductive materials used in the circuit tracks and exempts the conductive material used in the mounting of circuit components.

Table 1Mass of Cu to be etched away and Ag-ink required for different shares of conductive areas on the substrates utilized for the sensitivity analysis.

Conductive Area	Mass of Cu Etched Away from PCB [kg]	Mass of Ag-ink used in Printed Electronic [kg]
10% (baseline)	0.141	0.023
25%	0.117	0.057
50%	0.0784	0.115
75%	0.0392	0.172

3. Results

3.1. Environmental impacts for conductive materials

Since the fates at the EoL differ for the conductive and substrate materials, their life cycle impacts were calculated separately; Fig. 3(a) shows the climate change impacts from all the processes during the life cycle of the conductive materials used in PCBs and printed electronics for the 'without recycling benefits scenario'. For PCBs, the impacts from the conductive materials, i.e. Cu and solder, exceed the impacts from etching and WEEE recycling. The same phenomenon is observed for the printed electronic circuit, where the impacts of the Ag-ink and the Agbased conductive adhesive are higher than the impacts from printing and WEEE recycling. Another aspect evident from Fig. 3(a) is that the net impacts over the life cycle of Ag-based conductive materials in printed electronics (around 56 kg CO2 eq.) are significantly higher than those from the life cycle of Cu-based conductors in PCBs (around 4 kg CO2 eq.). Despite the quantity of Ag (0.01 kg) used for the conductive tracks in printed electronics being lower than that of Cu (0.015 kg) in the conductive tracks of PCBs (shown in Fig. 1 and Fig. 2), the much higher impacts of sourcing a precious metal like Ag also lead to higher conductor impacts in printed electronics.

The conductive adhesive (also utilizing Ag) is the largest contributor to the net impacts of the conductive materials in printed electronics. This high impact is attributed to the large quantity of adhesive considered in this assessment. Fig. 2 shows that the amount of conductive adhesive (0.177 kg) required in printed electronics is more than double the mass of solder (0.081 kg) applied for mounting components in PCBs. The quantity of conductive adhesive used is higher, as it represents an unoptimized amount because the mounting of the components in GREENSENSE was performed manually to some extent. In the initial phases of this study, the mounting process was excluded from the scope of the comparative LCA; however, the impacts of the conductive adhesive and the solder were later found to be pertinent.

The net impacts decrease significantly in the 'with recycling benefits' scenario shown in Fig. 3(b), where the recycled metals from the WEEE recycling are assumed to be circular and replace primary metals during the production of the PCBs and printed electronics. For example, the Cu etched away from the PCB surface is recovered and can replace Cu in the market; therefore the etching process offers an environmental benefit that is plotted along the negative vertical axis. Similarly, the solder in the PCB is an alloy of Sn, Ag, and Cu, all of which are recovered during WEEE recycling in their elemental forms (Reuter and Van Schaik, 2013), and the respective environmental credits are allocated to the WEEE recycling process for metal recovery. The WEEE recycling can also efficiently recover the Ag contained in the conductive tracks and adhesive of the printed electronics; when the environmental benefits of the recovery of Ag are accounted for in the assessment, the net impacts from the life cycle of the conductive materials in printed electronics decreases by a factor of 5, i.e. from 60 kg CO2 eq. in Fig. 3(a) to 12 kg CO2 eq. in Fig. 3(b). Although the net impacts over the entire life cycle of the conductive materials from printed electronics are still greater than those from PCBs, the impacts are more comparable and in a similar order of magnitude in Fig. 3(b) after the recycling benefits have been accounted.

3.2. Environmental impacts for substrate materials

Fig. 4(a) illustrates the climate change impacts over the entire life cycle of the substrate materials without considering any recycling benefits. Since the manufacturing of the FR4 for the PCB is an energy-intensive process (Nassajfar et al., 2021), the impacts from the PCB's substrate (around 100 kg of CO2 eq.) far exceed those from CNC-coated paper in printed electronics (around 35 kg of CO2 eq.). At the EoL in WEEE recycling systems, the epoxy content of the FR4 is combusted into fossil CO2, which is reported as the WEEE recycling impact for the PCB. In printed electronics, the combustion of both paper and CNC in metal

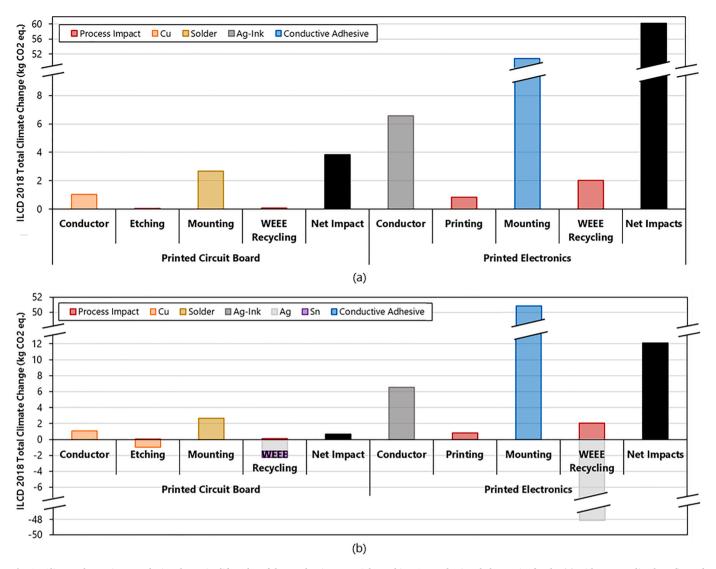


Fig. 3. Climate change impacts during the entire lifecycles of the conductive materials used in PCBs and printed electronics for the (a) without recycling benefits and (b) with recycling benefits scenarios; the individual impacts from the production processes on the horizontal axis (for example, conductor, etching, mounting, and WEEE recycling for PCBs) have been totalled up to represent the 'net impacts' over the entire life cycle.

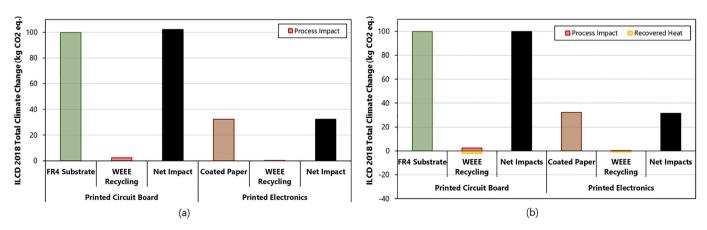


Fig. 4. Climate change impacts during the entire lifecycles of the substrate materials used in PCBs and printed electronics for the (a) without recycling benefits and (b) with recycling benefits scenarios; all the life cycle impacts for the substrate materials (for example, FR4 and WEEE recycling for PCBs) are totalled and represented as 'net impacts'.

smelters primarily produces biogenic CO2 (Muñoz and Schmidt, 2016), which is why the climate change impacts from WEEE recycling for printed electronics are lower than those for PCBs. Fig. 4(a) highlights that the CNC-coated paper substrates for printed electronics outperform the conventional FR4 substrates in PCBs in terms of environmental impacts by a significant margin. This result is surprising also when one considers that the impacts for the FR4 substrate have been optimized as the underlying dataset is for the industrial manufacturing of PCBs. On the other hand, the LCI modelled for the CNC-coated paper was based on lab- and pilot-scale processes; this implies that by scaling up the production of the CNC-coated paper there is a possibility of further reducing the impacts and enhancing the environmental edge of the paper-based substrate for printed electronics over the conventional FR4. Furthermore, paper with a 99% mass-share contributes less than 1% to the impacts from the final CNC-coated paper substrate (see Fig S1. in the Supplementry material.); instead, the CNC itself and the energy required to coat it on the paper contribute in total over 99% of the impacts. This underscores the possibility of further reducing the environmental impacts of the paper substrate by replacing the CNC, which is an energy-intensive nanomaterial to manufacture (Gao et al., 2018; Tao et al., 2017), with a low-impact coating material for paper that facilitates circuit printing using the Ag-ink.

The combustion of the epoxy, CNC, and paper in the metal smelters during the WEEE recycling generates heat. This heat replaces some energy input to the smelter and can therefore be considered as recovered heat depicted in Fig. 2 and accounted for in the 'lifecycle with recycling benefits' scenario. Fig. 4(b) shows that the energy recovery seems to offer minimal benefits over the entire life cycle: net impacts do not change significantly due to the energy recovery benefits as the production impacts of the FR4 and CNC-coated paper are (relatively) high.

3.3. Total environmental impacts and sensitivity analysis

From the previous chapters, it is evident that the conductive materials used in PCB have lower impacts than those from printed electronics, whereas the substrate of printed electronics outperforms the PCB counterpart in terms of environmental impacts. Thus, the net environmental impacts over the entire life cycle of the conductive materials and the substrate have been combined in this chapter to holistically compare PCBs with printed electronics. Additionally, the sensitivity of the net impacts to the conductive area is illustrated in Fig. 5(a) for the 'life cycle without the recycling benefits' scenario.

Fig. 5(a) highlights that despite this comparison being tipped in the favour of PCBs, the overall impacts from the substrate and conductive materials in the PCB slightly exceed those of the printed electronics

when the required conductive area is 10%. As the conductive areas increases, so do the impacts of the Ag-ink, and starting at 50% conductive area, the overall impacts from printed electronics exceed those from PCBs. Thus, printed electronics have an environmental advantage over conventional PCBs that is subject to the required conductive area. With an increase in the conductive area, the amount of Ag-ink to be printed on the substrate also increases (as seen in Table 1), and eventually, the impacts from this additional amount of Ag-ink compensate for and eliminate the benefits from the low impacts of the substrate material. Additionally, the significant contribution from the conductive adhesive to the impacts of printed electronics highlights that the unoptimized and manual mounting process is detrimental to the environmental performance of printed electronics.

Another goal of the sensitivity assessment was to compare subtractive and additive manufacturing processes: Fig. 5 maps the increase in the impact from the Ag-ink in printed electronics due to the increase in the conductive area. As the required conductive area increases, the amount of Cu that needs to be etched off the PCB decreases (also shown in Table 1), implying that lower amounts of chemicals and energy are required for the etching process. Thus, the net impacts over the lifecycle of the Cu tracks in PCBs decrease with an increase in the conductive area (this decrease however is imperceptible in Fig. 5 because of the low net impacts of Cu in PCB and can be seen clearly in Fig S2. of the Supplementry material.). This verifies that additive manufacturing is suitable when the manufacturing requires the addition of small quantities (Keskinen, 2012; Kunnari et al., 2009). When large quantities need to be added, such as in the case of 75% conductive area, subtractive manufacturing is more suitable considering the material constraints of the current additive technology for the manufacturing of printed electronics.

Finally, Fig. 5(b) compares the net impacts of PCBs with printed electronics for the 'life cycle with recycling scenario'. It can be seen that by ensuring the recycling of Ag or only using recycled Ag in the production processes, printed electronics have a constant sustainability edge over PCBs, irrespective of the required conductive area. Therefore, recycling printed electronics as e-waste is crucial to their environmental competitiveness. As evident from Fig. 3(b) and Fig. 4(b) WEEE recycling should be prioritized for printed electronics because it contributes little to the lifecycle impacts while generating significant environmental benefits from recycled materials.

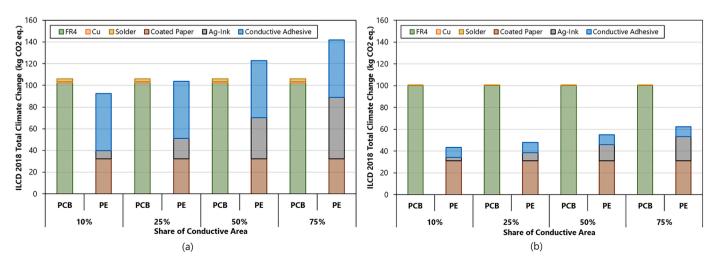


Fig. 5. Influence of the conductive area on the total climate impacts over the entire life cycle of PCB and printed electronics (represented as PE) (a) without recycling benefits and (b) with recycling benefits scenarios; 10% share of conductive area is the baseline for this study.

4. Discussion

4.1. Role of design for recycling

This study considers lifecycles with and without recycling to understand the significance of WEEE recycling on the sustainability of printed electronics. It is important to note that accounting for the recycling benefits as per the "avoided burden" approach as performed in the 'with recycling benefits scenario' is controversial: although ISO 14,040 instructs on accounting for the recycling benefits in an LCA, these benefits should be availed only once. However, if not properly monitored, both the recycler (in the first life cycle) and the user of the recycled product (in the second life) may rightfully claim these benefits leading to unjustified double counting in the LCA (Santero and Hendry, 2016). Moreover, it is possible to theoretically account for recycling benefits in an LCA even if the materials intended and sent for recycling never replace primary materials due to, for example, quality constraints; in such a case, waste reported to be recycled is not actually recycled and the theoretical environmental benefit is never realized. Thus the 'life cycle with recycling benefits' scenario is only used as a theoretical tool in this study to assess the relevance of treatment and recycling of printed electronics as WEEE at the EoL. Inefficiencies exist in the complex WEEE recycling and management systems (Evans and Vermeulen, 2021) that are captured in the realistic 'life cycle without recycling benefits'

Notwithstanding, recycling is crucial and the design of printed electronics should be in accordance with the design for recycling concepts (Van schaik and Reuter, 2012). As evident from the final results, recycling of printed electronics makes them environmentally viable as alternatives to PCBs; this is because it allows for the recovery of Ag, which is a precious and scarce metal with high sourcing impacts, from the printed electronic waste. The extensive use of and dependency on Ag is already known to be a sustainability challenge in the printed electronics community (Välimäki et al., 2020), and thus, recovery of Ag at the EoL is recommended (Kunnari et al., 2009).

Yet, due to the envisioned application of printed electronics primarily being limited to applications with short lifetimes and high market demand, there is a growing appeal to the development of biodegradable and compostable printed electronics (Hakola et al., 2021). The idea behind this approach is to avoid the environmental issues originating from the large quantities of persistent e-waste produced from "use and dispose of applications of printed electronics; many small devices with a wide market distribution making the collection at EoL for effective waste management tough (Zvezdin et al., 2020). The environmental challenge with composting printed electronics is twofold: the net environmental impacts over the lifecycle will remain high as precious Ag is basically lost to the environment and not recovered at the EoL; moreover, the nano-Ag is toxic and its release to the environment through the leaching of the waste may create ecotoxicological issues (Wiklund et al., 2021). Other unforeseen environmental issues may also arise with composting or the direct disposal of printed electronics to the environment; hence, biodegradability and compositing tests are required to assess the viability and environmental safety of compostable electronics (Zvezdin et al., 2020)

Specifically for paper-based electronics, paper recycling has also been successfully demonstrated as an EoL option to recover the paper from the substrates (Glogic et al., 2021). The challenge with considering paper recycling at the EoL is that Ag(https://doi.org/10.1080/09276440.2022.2128259) will probably be lost in the de-inking process step and its recovery is uncertain. Needless to say, adopting paper recycling at the EoL of printed electronics implies preferring the recovery of paper (a low-cost, renewable, and low-impact material), over precious Ag (with high sourcing impacts). A case study in literature (Hakola et al., 2021), showed that Ag in paper-based electronics has a significantly higher economic value than paper; thus the recycling of Ag from printed electronics is also economically preferable to recycling

paper. The future applications of printed electronics are anticipated to generate large volumes of waste containing Ag, making the economics of Ag recycling feasible and sustainable from printed electronics (van Beukering et al., 2014). Even recovering small quantities of precious metals from e-waste can be economical since the existing WEEE recycling infrastructure (Välimäki et al., 2020) can efficiently extract the metal from waste with low metal content. Considering these arguments and the results, the treatment of paper-based electronics as e-waste at the EoL and sending them for WEEE recycling seems to be the most sensible option. As long as the current printed electronics technology will be heavily reliant on Ag for conductive functionality, designing electronics to be compostable or ready for paper recycling seems to be unsuitable from an environmental as well as an economic perspective.

It is relevant here to also discuss and dispel some misconceptions in the minds of the developers of printed electronic systems about the waste management system. One common question is about the need for selecting a single EoL option out of the three: composting, paper recycling, and WEEE recycling; why can the paper-based printed electronic not be dismantled in a way to segregate the Ag-containing fraction from the paper and then recycling both fractions separately to recover Ag and paper? The reason for this is the current waste management system that relies on source segregation of compostable, paper, and electronic waste and aims to minimize the flow of materials between different waste streams after the collection. Therefore, segregating and recycling the paper fraction from (easy-to-dismantle and paper-based) electronic waste collected for WEEE recycling at the EoL is unlikely unless the WEEE recycler is given an economic incentive and the relevant connections with paper recyclers. Since the segregation methods employed in waste management have inefficiencies, a WEEE recycler may not wish to bear the risk of losing some valuable Ag in the paper fraction and therefore decide to simply send the paper-based electronic waste in its entirety to smelters so that the maximum value of Ag can be recovered. In the future, better coordination for segregation and feedstock exchange may exist between the recyclers and managers of the different waste streams. However, the present reality of the waste management system needs to be accepted as it only allows the sorting and treatment of paper-based printed electronics as one kind of waste (Bunge, 2012, 2018; Evans and Vermeulen, 2021; Singh et al., 2014). For the current waste management system, some printed electronics also pose logistical challenges associated with the collection of large quantities of smaller devices dispersed devices at EoL.

As printed electronics are still at an early stage of development, it is valuable to work with the developers to identify and implement ecodesign (J. Li et al., 2015) considerations in all phases of the life cycle including the EoL (Manjunatheshwara and Vinodh, 2021). Already identifying the need to focus on recycling for an early-stage technology, like printed electronics, can contribute significantly lowering the impacts when the technology reaches maturity (Kunnari et al., 2009); thus, it is necessary to have a circular vision for printed electronics (O'Connor et al., 2016; Zvezdin et al., 2020).

4.2. Legislative perspective

Besides the identification of the most suitable EoL treatment for printed electronics (with the support of the above LCA calculations), it is also relevant to see how the proposed novel applications for printed electronics and their EoL-pathways fit into the framework of the existing legislation. For example, the WEEE directive (European Union, 2012) of the EU clearly demarcates 10 categories of electrical and electronic equipment that should legally be recycled as WEEE, and that plastics and electronics fractions should be separated and recycled in the EoL process.

A medical device, such as the GREENSENSE biosensing platform, despite offering the possibility of being compostable at the EoL, would be legally classified as a 'medical device' and must be recycled with other e-waste at the EoL as per the WEEE directive. The use of a battery

in a printed electronic system makes the Battery Directive (European Union, 2006) from the EU applicable as well. According to this directive, the battery should be dismantlable from the device and it has to be removed at EoL for separate recycling. Thus, designing any printed electronic system with a power source for biodegradation in nature would likely infringe the WEEE and battery directives and the developers of the printed electronic systems need to be mindful of this.

Nevertheless, novel applications are being envisioned for electronics that are either beyond the scope of existing legislation or may conceptually fall under the scopes of multiple legislation. The latter is particularly the case when printed electronic systems are utilized to impart electronic functionality to non-electronic applications such as clothing and packaging. Smart packaging and smart textiles are such classes of general products with add-on electronic functionalities made possible by printed electronics (Glogic et al., 2021; Keskinen, 2012; Kunnari et al., 2009). The management of smart packaging and textiles at the EoL is hampered due to technical challenges, for example, those associated with the recycling of textiles containing electronic components, as smart textiles are not classified as e-waste under the existing WEEE legislation. Similarly, smart packaging contains electronic components but cannot be classified as WEEE and has to be recycled as packaging waste under the packaging waste directive of the EU (European Union, 1994) even though the electronics in the smart packaging affect the quality of recycled plastics (Keskinen, 2012). Hence, the legislation need updates and amendments to establish a framework for the management of these novel hybrid applications containing electronics (Veske and Ilén, 2020). Without the necessary updates, antiquated legislation may further lead to the exacerbation of the waste problem: technological developments in the printed electronics sector may create new and difficult to manage waste streams.

5. Conclusions

Based on the comparative carbon footprint analysis presented here, printed electronics offer an advantage over conventional PCBs, especially for circuits requiring a small conductive area. These results are valid despite the models for the printed electronics being based on unoptimized lab- and pilot-scale processes and the comparison being biased in favour of PCBs. The sustainability edge of printed electronics over PCBs is greater and consistent when WEEE recycling is undertaken for the former at the EoL to recover the precious Ag. Many varieties of printed electronics have been designed to be compostable and biodegradable for the sake of eco-friendliness. However, such EoL options will hamper the net sustainability over the entire life cycle of printed electronics as long as Ag is the key ingredient in conductive substances of printed electronics. Furthermore, developers should be mindful of the current legislation as the development of compostable electronics may infringe legislation such as the WEEE directive. For paper-based electronics, design for e-waste recycling is crucial and it offers a path to lowimpact electronics that are sustainable alternatives to conventional PCBs.

Funding

This project has received funding from the European Union's Horizon 2020 research and innovation program under grant agreement No. 761000 GREENSENSE.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The data that has been used is confidential.

Acknowledgments

The authors acknowledge the expertise and the data provided by Dr. Valerio Beni and his team at RISE during this project. The authors also acknowledge that the suggestions from Dr. Dirk Hegemann at Empa on the behaviour of nanomaterials were important in this study. Finally, the authors express their gratitude to Dr. Sandra Martinez and her team at LEITAT Technological center for their valuable feedback, encouragement, and support.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.resconrec.2022.106757.

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