

Additively Manufacturing Printed Circuit Boards with Low Waste Footprint by Transferring Electroplated Zinc Tracks

Corresponding Author: Dr Jonathon Harwell

This file contains all editorial decision letters in order by version, followed by all author rebuttals in order by version.

Version 0:

Decision Letter:

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Dear Dr Harwell,

Thank you for submitting your manuscript, "Zero Waste Microelectronics and Biosensors Enabled by Overmould Printing of Bulk Zinc Tracks", to Communications Materials. It has now been seen by 3 referees, whose comments are appended below. You will see that while they find your work of potential interest, they have raised substantial concerns that must be addressed. In light of these comments, we cannot accept the manuscript for publication, but are interested in considering a revised version that addresses these serious concerns.

If any of the reviewer reports contain recommended references to be included in your manuscript, please check them carefully and only include those you think are relevant and help improve your manuscript.

We hope you will find the referees' comments useful as you decide how to proceed. Should further experimental data or analysis allow you to address these criticisms, we would be happy to look at a substantially revised manuscript. However, please bear in mind that we will be reluctant to approach the referees again in the absence of major revisions. If the revision process takes significantly longer than twelve weeks, we will be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Materials or published elsewhere in the meantime.

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When submitting your revised manuscript, please include the following:

-A response letter with a point-by-point reply to each of the referee comments and a description of changes made. Please include the complete referee report in the response letter. Please note that the response letter must be separate to the cover letter to the editors.

-A marked-up version of the manuscript with all changes to the text in a different colored font. Please do not include tracked changes or comments. Please select the file type 'Revised Manuscript - Marked Up' when uploading the manuscript file to our online system.

-A clean version of the manuscript. Please select the file type 'Article File'.

Please ensure that the following requirements are met, and that any relevant checklists are completed and uploaded under the 'Related Manuscript file' type with the revised article.

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Please do not hesitate to contact me if you have any questions or would like to discuss the required revisions further. Thank you for the opportunity to review your work.

Best regards,

Rona Chandrawati, PhD
Editorial Board Member
Communications Materials
orcid.org/0000-0002-9780-8844

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The article describes an original way to pattern Zinc traces on different eco-friendlier substrates. The article in general is well written with detailed explanations on the process and some demonstrations of the technology, those on sensors not really bringing added value. There are although a few little and more important things to revise therefore a major revision is required.

Nomenclature, formulation and presentation:

Title and in the text:

Overmould printing

This is an abuse of language to use the word printing when there is no ink involved. The formulation transfer printing should have never been used in the literature for such a process involving the transfer of solid structures. I invite therefore the authors to review their title and change it with a terminology representative of the process reported and work achieved.

In general the title is overclaiming in comparison to the achievements reported in the article.

Zero-waste: There is no strategy to recover the zinc, the substrate, only electronic components are recovered. Is this therefore really zero-waste? There are other works showing fully recyclable PCBs with proofs of recovery for all materials, and even reused. Here there are some claims but not backed up by concrete results.

Microelectronics: Microelectronic means design of silicon or other (SiGe, GasAs, etc) foundry-based components, this is PCB technology that is presented.

Biosensors: There is no biosensor reported in this paper, only speculations, these are touch sensors. A biosensor is a sensor that uses biological elements in its operation, enzymes, cells, etc.

In conclusion, the Title has to be fully reviewed.

Abstract

Line 4: As existing circuit designs, it is not clear what here circuit designs means, circuit designs refers to IC technology, not to PCB technology, this should be reformulated.

Line 8: Zinc, the most desirable degradable conductor... It is not clear why zinc is the most desirable degradable conductor. Zinc is a heavy metal, there are standards in relation to the quantity that can be released in soil/composting. Please review formulation or justify why it is the most desirable.

Line 10: Overmould printing is used, to be changed, also in the rest of the manuscript.

Line 11: Passive sentences Thus enabling fully degradable and biocompatible electronics, as well Line 18, Thus providing, the grammar should be reviewed

Layout not uniform and well formatted for the references, therefore to revise.

Section 1: Background

Line 68: Overmould printing is used why in line 77 the process is appropriately described as a transfer of zinc film to a target substrate by moulding the substrate material. Basically, this is a transfer process by moulding, there is no printing involved. Also the section 2 is named Transfer plating process, which corresponds actually to what the process is about. Perhaps change everywhere Overmould printing by Transfer plating process?

Line 69: Creating PCBs that are zero-waste, at the end of Life... Evidences are missing that the PCBs fabricated are zero-waste. Zinc and substrates are not recovered, or degraded in soil and compost. Basically, only the silicon components are

recovered, see my comments below for the related section 4.3.

State of the Art: There are some papers reporting on bulk transfer of Zinc conductors. The authors should review the state of the art and position their technology according to that approach as well. Notably check : <https://doi.org/10.1038/s41598-022-12162-z>

Section 2: Transfer plating process

The adhesion of this Zn seed layer is approximately 1 kPa, in shear, normal strength? Approximately should be replaced by an average value with standard deviation number coming from measurements of more than 1 sample, $n=3$ would be acceptable, 5 even better.

Line 151: Zn tracks embedded into a resin substrate: Different materials have been demonstrated, can all be called a resin? I think it could be good already for the reader at this stage to have an idea of the types of materials demonstrated.

Line 160: 5 μm was the smallest linewidth that could be resolved... What about the minimum/maximum thickness and especially the aspect ratio achievable, at 5 μm line width how thick can be the structures?

Section 3: Results

Lines 167-168: Major advantage of our method over other additive processes is almost independent of the area... This argument is not reasonable, in a roll to roll process few meters/seconds can be processed, how would you proceed with the given technique to pattern 30 meters, 1 km of web? So scaling up the method becomes non competitive (without talking about all the extra steps such as lithography, etc).

Figures 3 b, c, d, are not readable, too small figures characters, etc. My recommendation would be to put them below Figure a) on the same row, next to each other, to align them horizontally, not vertically.

Lines 214-215: The way the results are presented requires improvement so we can really understand how reproducible is the process and the conductivity value claimed. There should be a deviation of the thickness measurement, the number of samples measured given, resistance + thickness, and this number should be at minimum 5 (from same and also for 2 other batches of samples fabricated for comparison). And finally this must lead to an average conductivity value with a standard deviation.

And important, in the supplementary document, the summary table, the conductivity number cannot be rounded to be $2.0 \times 10^7 \text{ S/m}$, a value even higher than bulk value, there should be the average number value + standard deviation for $n=5$ at least.

Line 244: 1000 bending cycles at 9 mm of radius. What does happen at 8 mm, it does not reach 1000 cycles?

Lines 252-253: the measured resistance in comparable, quantitative values would be appreciated, how much difference in percentage, how many samples measured. Aging tests involve statistics, a minimum of samples should be tested, an average value, standard deviation and comparison then provided.

Figure 4 b, c, small, and hard to read as well.

pH should be written with a small p, not PH, to review all over the text.

Section 4: Implementation of real-world applications

Section 4.1: Touch/Sweat sensors. There is no sweat sensor reported, 2 touch sensors are presented, basic demos. Title has to be changed, formulations in the text and discussion with references to sweat sensors, biosensors, etc, should be removed, notably lines 321-322. There is no result showing a biosensor, and no results showing that the technology can be used as biosensors, wearables, etc. The 2 touch sensors show that 2 transducing principles are applied, but at the end, these are nothing more than two zinc electrodes used to measure a capacitor and a resistor.

Capacitive touch sensor: polymers, etc are humidity sensitive, what was the measurement during the measurement, and what will happen to the capacitance value at different relative humidity?

Lines 331-333: What was the power required to reach temperatures above 100°C , how stable was the heater in terms of drift of the resistance over time? How many samples tested, TCR value error, deviation for $n=5$ samples?

Section 4.3 "Fully degradable" Microelectronics...

Again quote "" is used, so what does mean exactly Fully degradable? Is it or not fully degradable, it is not proven in any case.

Section 4.3 "Fully degradable"

What degradable does mean here ? By which means, in soil, compost? Here it is dissolved. There is no recovery of zinc, or

substrate material, no clear therefore what no waste means, zinc, there is also silver, etc. where do they go? There is no proof that the zinc, silicon can be degradable in soil or be compostable. Reference 37, silicon is bioresorbable, degradation in soil or compost needs to be proven. All this discussion should be reviewed and reformulated since speculative.

Section 5: LCA

The paper claimed notably that the method is better than printing zinc in terms of conductivity, linewidth, stability, etc. Then it would be of high interest to have the LCA between the method reported and printed zinc, especially due to the fact that the method proposed involves more steps, substrate formation, transfer, some chemicals, lithography, etc.

Section Methods:

Materials (section should be created)

Nothing is said about the substrates materials, a section should be added on the substrates, materials used linked to suppliers. It is not clear how PHBV actually is applied, pellets from which supplier, etc, same thing for the other materials. More details should be given so the readers can reproduce the procedures.

Heater/Temperature sensor measurements

Mention the model of Thermal camera used. Tape was used to glue sample on metal block, is it a thermally conductive tape? What about the emissivity value used to measure the temperature on the heater, how was done the calibration?

Supplementary information

SI13. Stress testing

Title is not appropriate, it is not a stress test of the film, it is an adhesion test.

Titles of tables should be above the tables, not below.

Table S1: there is a \$ sign for scarcely available materials, I was not able to spot that sign in the table information.

Table S2 has its caption on the other page. More important, the conductivity value included needs to be reviewed and comparison with laser patterned zinc layer transfer added in the comparison.

Reviewer #2 (Remarks to the Author):

Dear Authors,

Thank you for this comprehensive and interesting work. I particularly appreciate the fact that you incorporated LCA in the study instead of just claiming sustainability.

While the manuscript is solid and written in clear language, some concerns need to be addressed before I can recommend this study to be published. Please consider my comments:

1. You mention the issue of e-waste and in parallel discuss the applications like smart packaging. However, those single-use devices are unlikely to be treated as electronic waste and will most probably end-up in the municipal waste stream. This gives a strong motivation for designing the device in a biodegradable way, while for most other scenarios recycling would be the most desirable EoL option.

2. You propose the usage of Chitosan as a substrate material as well as "adhesive" layer between the Zn electrodes and the paper substrate. Especially in capacitive applications I see some issues with that: the electrical properties of Chitosan are highly dependent on ambient humidity levels, the resistance follows an exponential curve with the largest change in the lower humidity range, as described in:

Zikulnig, J., Lengger, S., Rauter, L., Neumaier, L., Carrara, S., & Kosel, J. (2022). Sustainable printed chitosan-based humidity sensor on flexible biocompatible polymer substrate. *IEEE Sensors Letters*, 6(12), 1-4.

Another recent source dealing with the electrical properties of Chitosan:

E. E. Netey-Oppong, R. Muhammad, E. Ackah, H. Yang, A. Ali, H.-W. Jeong, S.-W. Kim, Y.-S. Seok and S. H. Choi, "Development of a Sustainable Flexible Humidity Sensor Based on Tenebrio molitor Larvae Biomass-Derived Chitosan," *Sensors*, vol. 25, no. 2, p. 575, 2025

Considering this, your application examples of the IDE sensors would not work reliably. In general, to guarantee stable operation of electronic devices, an inert substrate material should be preferred to avoid cross-talk.

3. For most devices, especially in the field of printed electronics, the substrate material accounts for the largest part of the

mass. Historically, this was somewhat misleading for researchers to assume that the focus on the substrate reduces the environmental impact the most, while recent research has shown that silicon components as well as silver inks pose as environmental hotspots:

Zikulnig, J., Carrara, S. & Kosel, J. (2025). A life cycle assessment approach to minimize environmental impact for sustainable printed sensors. Scientific Reports.

Can you please elaborate that further in the context of your work?

4. Please describe the resist stripping process. Does it affect the Zn layer and why (not)?

5. On page 2 you describe that degradable metals have an “awkward” melting point – I don’t think this wording is appropriate, it sounds like slang.

6. The IDE resistive touch sensor: It is perhaps inaccurate to say that skin is conductive because of sweat – it is rather because of salts and impurities present in the skin and the impedance is highly dependent on the frequency. Please correct that.

7. For temperature sensing, you mention a change in resistance of 0.7 Ohm/degC. Please express that in terms of TCR and compare with Zn bulk metal.

8. Thermal imaging: Please provide information on the thermal emissivity (epsilon) used for temperature measurement.

9. Why are you using wirebonding? Flip-chip bonding could reduce the material needed for chip integration and with the high resolution of your method, replicating the chip footprint should not be an issue. Please also briefly discuss the issues of thermal processing with temperature sensitive substrate materials, like PLA, Chitosan, Cellulose Acetate.

10. I have an issue with the EoL of the biodegradable sensor. How can the GWP be negative, when it degrades? Usually, under biodegradation, the CO₂ stored in the material is released. Please elaborate on this further!

11. FR-4 substrates contain flame retardants. How would you mitigate the issue of flammability with your devices?

Reviewer #3 (Remarks to the Author):

The manuscript presents an overmould printing method for fabricating fully degradable electronic circuits using bulk zinc tracks embedded into biodegradable substrates. This approach enables high-resolution patterning, near-bulk conductivity, and compatibility with a wide range of substrate materials. The authors demonstrate various devices including touch sensors, heaters, and microelectronic circuits, and provide a life-cycle analysis showing reduced environmental impact compared to traditional FR4 PCBs. The method is promising for sustainable electronics and low-cost biodegradable IoT systems. However, several issues should be addressed before the manuscript can be considered for publication:

1. The manuscript lacks quantitative data on fabrication yield and reproducibility across substrates and pattern geometries. It is unclear how robust or scalable the process is in practice.

2. The long-term operational stability of the devices is not sufficiently evaluated. While bending and humidity resistance are shown, extended testing under realistic conditions is needed to assess reliability.

3. The comparison with commercial copper-PCBs is overstated. Direct benchmarking of full circuit performance, including thermal limits, signal integrity, or integration density, is absent.

4. Recommend citing ACS Nano 2021, 15, 9328; Nat. Commun. 2022, 13, 3223; TrAC, Trends Anal. Chem. 2021, 143, 116369; and TrAC, Trends Anal. Chem. 2018, 102, 236 in the Introduction to better contextualize recent developments in degradable microelectronics and biosensors.

5. Known limitations of the method (e.g., thermal sensitivity of substrates, Zn oxidation, adhesion failures on certain materials) are not discussed, which hinders realistic evaluation of the technology's scope.

6. Key datasets such as sheet resistance, temperature response, and degradation time are presented without error bars or information on measurement variability. Statistical analysis is needed to validate claims.

7. The recycling demonstration is qualitative. Quantitative data on component recovery efficiency, functional performance after reuse, and degradation of parts during disassembly would greatly strengthen this part.

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Version 1:

Decision Letter:

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Dear Dr Harwell,

Thank you for submitting your revised manuscript, "Zero Waste Microelectronics and Biosensors Enabled by Overmould Printing of Bulk Zinc Tracks", to Communications Materials. It has now been seen again by the 3 referees, whose comments are appended below. You will see that Reviewer 1 and 3 continue to raise substantial concerns. In light of these comments, we cannot accept the manuscript for publication but are interested in considering a revised version that addresses these serious concerns.

We hope you will find the referees' comments useful as you decide how to proceed. Should additional data or analysis allow you to address these criticisms, we would be happy to look at a revised manuscript.

To allow us to move forward with your work, we also ask that you edit your manuscript according to the attached table.

Please read this document carefully as we will be unable to further assess your revised paper until these important points are addressed.

Please outline all revisions made in the right-hand column and return the completed table with your updated manuscript files as a Related Manuscript file.

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- A clean version of your revised manuscript with no mark-ups
- A marked-up version of your paper with all changes highlighted in a different colour

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We hope to receive your revised paper within twelve weeks, but we understand that revisions may take longer. Please let us know if you find that the revision process will take substantially more time.

We are committed to providing a fair and constructive peer-review process. Please do not hesitate to contact me if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

Rona Chandrawati, PhD
Editorial Board Member
Communications Materials

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

Reviewer 1

The authors made large efforts to improve significantly the paper. There are still a few points to be addressed before acceptance for publication.

Biodegradation of Zinc PCB. I think the numbers and process to reach the conclusion of 99% biodegradable can be questioned according on how you consider what is safe, not safe, degrades or compost. Because of this I would:

- Remove 99% in the title. It can appear elsewhere, but this is claim is incomplete, not considering all the aspects, and therefore could be debated.

- Saying that zinc is safe once degraded should be removed as well. Zinc is a heavy metal and limited in soil content. I believe that following the composting regulations the quantity of zinc would be too much (we did the test) and therefore the PCB would not be compostable. It is easy to say everything degrades in soil, but in practice it is not applicable. I agree that the PCB biodegrades in the conditions given, but still care should be taken about that this is safe (answer to comment 8 says that Zn safely degrades), release benign products, etc.

Comment #3:

Indeed, the comment asked to precise in the introduction the statement mentioned in the abstract, to justify it, why zinc is the most desirable degradable conductor.

However, in the information added. I do not understand why emphasis is put on biocompatibility, we are talking about degradable conductors in the environment, and therefore the discussion should be not limited to the fact it is biocompatible (this means with human body and also in a limited daily quantity if bioresorbed), listing and comparing with sodium and calcium, that are not really adapted for making electronic boards (iron not ideal, but could be considered). W, Mo, Mg are missing in the discussion for instance.

There is also an issue with reference 21 which refers to PEDOT:PSS. I believe this reference or numbering should be changed. And I would really like to have a look at this reference to see what the conclusions are and they were obtained.

Comment #26:

All papers listed to backup the following discussion mainly address clinical and in body applications. We have published different papers on zinc and transient electronics and we look at the literature. Everyone claims and refer to some paper that Zinc degrades well in soil, etc, but looking at all references and mechanisms, they are related to implantable, in body, in liquid degradation (Such as PBS). So what is described below and the references given are for zinc in body. What about zinc outside the body, have you found any references on studies looking at zinc degradation in soil ? We have not. In the text below mechanisms are given, etc, and soil is listed among other environments, this requires proper references if you want to claim that zinc degradation has been studied demonstrated in soil.

Zn belongs to a family of 'degradable metals' (alongside magnesium, molybdenum and iron), offering intermediate corrosion rates. There have been several reviews that confirm Zn's biocompatibility, and its appropriate degradation into naturally occurring materials such as zinc oxide, zinc carbonate, or zinc phosphate in water, soil or the human body (

Comment #15:

Thank you for the new experiments made on the conductivity of the Zn films, but I have a serious issue still with the value reported which for me questions the robustness of the methodology used. How can you reach conductivities higher than the bulk zinc value ? How can you explain this ? And this value is still now referenced in the comparison table.

Comment #24:

I have also serious concerns on the methodology used to measure the TCR coefficient (and therefore the temperature in the experiments), since the TCR value found is 50% higher than the bulk value, how can this be physically possible ?

Comment #29:

How was selected this emissivity 0.95, was it measured ? From literature ? Did you measure temperature on substrate or on zinc track ?

Reviewer #2 (Remarks to the Author):

Dear Authors,

Thank you for addressing my comments and performing a substantial review which has significantly improved the quality of the work. I recommend the paper to be published in this form.

Reviewer #3 (Remarks to the Author):

The authors have made a substantial effort to address the reviewers' concerns. The revised manuscript includes additional data on fabrication yield, stability, and recycling, which improves the overall presentation. However, several critical issues regarding experimental rigor and data reliability remain unresolved. In its current form, the manuscript still does not meet the standards for publication. The following points require attention:

1. Data presentation remains weak. Several key images (e.g., Figures 1f, 3a, 9e) are blurred or affected by glare, which diminishes their evidentiary value. Figure composition should be improved to ensure clarity and professional presentation.
2. Yield data are provided only for UV resin substrates. No yield or reproducibility analysis has been reported for the more relevant biodegradable substrates (e.g., paper, PHBV), which undermines claims of process robustness. In addition, the overall writing quality needs improvement, as the manuscript still contains numerous basic errors that affect readability.
3. The scalability claim is supported only by a 15 × 30 cm visual demonstration. Quantitative mapping of thickness and sheet resistance across large areas is needed to substantiate uniformity and reproducibility.
4. Evidence for benign degradation remains insufficient. Visual soil tests alone cannot prove safety. Chemical analysis of degradation products (e.g., ICP-MS, SEM-EDX) and ecotoxicity validation (e.g., seed germination) are required.
5. Comparisons to FR-4 PCBs are overstated. Literature tables (Table S4) are not a substitute for direct benchmarking. High-frequency signal integrity tests on the fabricated PCBs remain essential.
6. Heater stability was tested only for 60 minutes. No thermal cycling experiments were performed to evaluate long-term reliability under repeated stress, which is a key failure mode.
7. Recycling experiments show functional component recovery, but recovery efficiency is not quantified. Reporting the percentage of successfully retrieved components is critical for evaluating circularity.
8. Several plots central to degradation claims (e.g., Figure 9c) are presented without error bars. Proper statistical treatment with multiple trials ($n \geq 3$) is necessary to validate the reported trends.
9. Humidity tolerance is reported only qualitatively. Quantitative assessment of circuit performance under varying humidity (e.g., standby current, noise levels) is required to establish operational limits.

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Version 2:

Decision Letter:

Dear Dr Harwell,

Thank you again for submitting your manuscript "Additively Manufacturing Printed Circuit Boards with Low Waste Footprint by Transferring Electroplated Zinc Tracks" to Communications Materials. We have now received reports from 2 reviewers, which are copied below. Based on their comments, we are delighted to accept your paper for publication.

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We look forward to publishing your paper.

Best regards,

Rona Chandrawati, PhD
Editorial Board Member
Communications Materials
orcid.org/0000-0002-9780-8844

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

Reviewer 1

I am in general fine with the modifications performed and complementary information provided. When I was saying no paper on zinc in soil, I meant degradable electronics decomposed in soil passing composting conditions. For your information, aluminum on paper PCBs used in RFID and even some companies using printed Ag as metallisation have succeeded to be compostable (us with Zinc we failed). Finally, it is still not clear to me how you can achieve with electroplating bulk conductivity. I wish you lot of citations !

Reviewer #3 (Remarks to the Author):

The authors has responded the comments.

***As a new journal, we would greatly appreciate any comments you have about your experience at Communications Materials. I hope that we have been able to meet your expectations and look forward to working with you again in the future.

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We thank the reviewers for their in-depth reviews and present the renewed article with the following changes to address the reviewer's concerns.

All text and figure changes have been highlighted and directly noted in these responses. However we would also like to point out that these changes have required us to change the ordering of the sections and also split the figures up in order to preserve the flow of the manuscript.

Reviewer #1 (Remarks to the Author):

1. The article describes an original way to pattern Zinc traces on different eco-friendlier substrates. The article in general is well written with detailed explanations on the process and some demonstrations of the technology, those on sensors not really bringing added value. There are although a few little and more important things to revise therefore a major revision is required.

Title and in the text: Overmould printing; This is an abuse of language to use the word printing when there is no ink involved. The formulation transfer printing should have never been used in the literature for such a process involving the transfer of solid structures. I invite therefore the authors to review their title and change it with a terminology representative of the process reported and work achieved.

.....In conclusion, the Title has to be fully reviewed.

We understand the reviewers concerns with the wording used here, and so we have changed the title to ***“Designing for End-of-Life Electronics: Electroplated Zinc Conductive Tracks enabling 99% degradable High-Performance PCBs”***

This removes the problematic references to printing, microelectronics and biosensors, and replaces the “zero waste” with “99% degradable”. This calculation is discussed later in the Reviewer 1, Comment 26.

2. Abstract

Line 4: As existing circuit designs, it is not clear what here circuit designs means, circuit designs refers to IC technology, not to PCB technology, this should be reformulated.

After reviewing the abstract, we have fully modified this to reflect the main strengths of the paper and the reviewers comments. The new abstract is as follows:

Recyclable, degradable, and printed electronics are gaining attention as solutions to the growing e-waste problem, but achieving high-resolution, high conductivity circuits with low-waste end-of-life (EoL) remains difficult. We present Printed Circuit Board (PCB) assemblies where 99.7% of mass degrades into benign EoL products in controlled or soil environments, supporting circularity for integrated circuits (ICs). Our “growth-and-transfer” process electroplates conductive bulk zinc (Zn) onto a sacrificial carrier, then transfers it to a biodegradable substrate. This yields 5 µm track resolution, 3 mΩ/sq sheet resistance, and a shelf life over one year. PCB performance, lifetime, and degradability were tested, showing rapid degradation in soil under composting conditions. Life Cycle Assessments (LCAs) benchmarked against conventional FR-4 copper PCBs reveal a 79% reduction in Global Warming Potential. The approach also enables easier recovery of high-value components such as ICs and LEDs for recycling by rinsing in a mild acetic acid solution, causing minimal damage to critical components. By combining high-performance circuitry with rapid degradation and reduced environmental impact, this method offers a potential solution to the growing electronic waste. It can significantly lower lifecycle impacts, improve component recovery, and ensure that, if disposed of in landfills, PCBs degrade into harmless end products.

3. Line 8: Zinc, the most desirable degradable conductor... It is not clear why zinc is the most desirable degradable conductor. Zinc is a heavy metal, there are standards in relation to the quantity that can be released in soil/composting. Please review formulation or justify why it is the most desirable.

We appreciate the reviewers comment; however, a detailed discussion on why zinc was chosen does not fit into the abstract, so therefore we have softened the wording in the abstract. To satisfy the reviewer, we have added a further discussion to justify why Zinc was as follows in Section 1, line 58: *“Among the degradable metals, zinc (Zn) is generally considered to be the most suitable because it has the best balance between abundance, biocompatibility^{22,23}, conductivity, and stability^{24,25}. Zn is a relatively abundant element that has an environmental footprint ~ 100x lower than Ag²⁶. It is an essential trace element that is required by the human body in quantities almost as high as iron, and it degrades rapidly in soil to benign and naturally occurring compounds such as ZnO, ZnCO₃, or Zn₃(PO₄)₂ depending on the soil composition²¹. In addition, it has a high conductivity of 1.7×10^7 S/m, and it is much more resistant to environmental degradation than other biocompatible metals such as iron, sodium, or calcium. Despite these advantages, manufacturing PCBs using Zn has proved to be especially challenging.”*

4. Line 10: Overmould printing is used, to be changed, also in the rest of the manuscript.

As discussed in the first comment around the title, we have swapped all references to “overmould printing” with “Growth-and-Transfer process.” This reflects the fact we use electroplating before transferring to the biodegradable substrate.

5. Line 11: Passive sentences Thus enabling fully degradable and biocompatible electronics, as well Line 18, Thus providing, the grammar should be reviewed

As discussed above, the abstract has been fully reworded

6. Layout not uniform and well formatted for the references, therefore to revise.

We have checked the references again and these have been updated to fit with nature publishing group’s referencing style. We apologise for this inconvenience.

7. Section 1: Background. Line 68: Overmould printing is used why in line 77 the process is appropriately described as a transfer of zinc film to a target substrate by moulding the substrate material. Basically, this is a transfer process by moulding, there is no printing involved. Also the section 2 is named Transfer plating process, which corresponds actually to what the process is about. Perhaps change everywhere Overmould printing by Transfer plating process?

We agree with the reviewer; as discussed earlier, we have changed all references to overmould printing to “growth-and-transfer”.

8. Line 69: Creating PCBs that are zero-waste, at the end of Life... Evidences are missing that the PCBs fabricated are zero-waste. Zinc and substrates are not recovered, or degraded in soil and compost. Basically, only the silicon components are recovered, see my comments below for the related section 4.3.

The discrete component can be recovered and reused (this is shown in the article); however, many of these components can also be manufactured to be degradable, as supported by evidence in the literature.

To prove degradation of the substrate and Zn tracks, we have performed degradation experiments of two weeks under composting conditions (70 °C, 85 % RH) of circuits based on paper/chitosan or PHBV, and have added the below images to the SI.

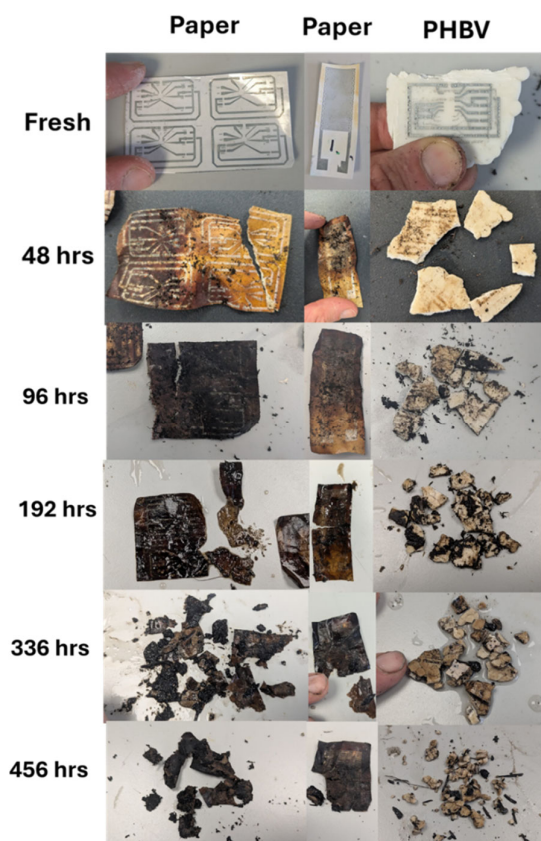


Figure S21 – Degradation images of circuit samples based on either paper or PHBV substrates before and after being buried under soil at industrial composting conditions (70 °C, 85 % humidity)

We agree that ‘zero waste’ is probably not suitable. However, as we discuss later in reviewer 1 comment 26, we have estimated that, 99% of the PCB mass is degradable to benign end products, so we have used this terminology where previously we said ‘zero waste.’

Line 87 now reads: “thus creating PCBs where more than 99 % of their mass can safely degrade at their end-of-life (EoL). “

Line 100 now reads: “that have minimal impact at their EoL”

Line 292 now reads: “4.2. Towards zero-waste”

Line 296 now reads: “degrade to non-hazardous products at their EoL”

Further discussion of degradation is provided in the comments to **Reviewer 1 in Comment 26**

9. State of the Art: There are some papers reporting on bulk transfer of Zinc conductors. The authors should review the state of the art and position their technology according to that approach as well. Notably check : <https://doi.org/10.1038/s41598-022-12162-z>

We thank the reviewer for pointing this out. We have added a few new citations to satisfy the reviewer and have added a discussion of this work into the introduction (line 71) as follows:

“For instance, Gopalakrishnan et al made biodegradable chipless sensors by laser cutting commercial Zn foils into traces of 1 mm width, and then sandwiching them between two sheets of polylactic acid (PLA) to make a durable but biodegradable sensor²⁹. This method achieved excellent conductivity, stability, and biodegradability, however the resolution was limited to ~1 mm due to the mechanical fragility of the foil when cut into finer features. This low resolution limits the potential applications, and so research groups have since explored additive printing processes from metal nanoparticle inks in order to improve upon the resolution and also minimise wasted material from the subtractive cutting process. ”

10. Section 2: Transfer plating process.

The adhesion of this Zn seed layer is approximately 1 kPa, in shear, normal strength? Approximately should be replaced by an average value with standard deviation number coming for measurements of more than 1 sample, n=3 would be acceptable, 5 even better.

The adhesion is defined in terms of normal strength, measured using a weight hanging setup as discussed in the SI. We have altered the wording of the statement to clarify this. In addition, we have repeated the measurement to n=5 samples and have updated the value and the data on figure S8 accordingly. Note that we do not include error bars in the value for the adhesion of the Zn to the new substrate because our setup is unable to measure adhesion values higher than this, and so the value of 500 kPa represents a lower limit rather than a real data point. However, the key takeaway from this measurement is that the adhesion to the new substrate is orders of magnitude higher than the adhesion to the Al carrier – remains unaffected by this.

The statement on line 122 now reads

“Crucially, the normal adhesion strength of this Zn seed layer to the Al carrier is 1.6 ± 0.6 kPa (n=5 samples -see figure S7&8 for measurement),”

11. Line 151: Zn tracks embedded into a resin substrate: Different materials have been demonstrated, can all be called a resin? I think it could good already for the reader at this stage to have an idea of the types of materials demonstrated.

We had chosen to discuss only UV resin at this point in order to keep the discussion concise.

However, we accept the reviewer’s point that this can make it confusion for the reader to know what materials are possible. We have adjusted the paragraph at line 168, and it now reads as follows:

“To assemble an electronic circuit, the Zn tracks are transferred from the Al carrier substrate to a new and insulating substrate. This is achieved either by pouring a liquid resin over the tracks, melting a thermoplastic over the top and allowing it to solidify, or by pouring a solution of a biopolymer dissolved in a solvent (e.g., cellulose acetate in acetone) and letting the solvent evaporate. Upon solidification, the high surface area of the rough Zn surface enables strong adhesion between the Zn and the new substrate. In the case of UV resin, the adhesive strength of this bond exceeds the measurement limit of our setup (500 kPa), indicating it is at least two orders of magnitude stronger adhesion than between Zn and the Al carrier substrate. Hence, the new substrate material can be peeled away to leave a clean Al substrate with the Zn embedded inside it as shown in Figure 1(f) and supplementary video 1. The Al sheet can then be re-used for further plating processes, reducing material waste from the production process. The final result is a high-quality PCB made from bulk Zn tracks embedded into the chosen substrate. .”

12. Line 160: 5 um was the smallest linewidth that could be resolved... What about the

minimum/maximum thickness and especially the aspect ratio achievable, at 5 μm line width how thick can be the structures?

Zn thicknesses up to 20 μm can be grown before delamination of the Zn film starts to occur, but the key determinant of the aspect ratio of the Zn tracks is limited by the thickness of the photoresist used (in our experiments). Given that our photoresist thickness mentioned in the paper is $\sim 5\ \mu\text{m}$, this limits our aspect ratio to 1:1 for 5 μm patterns. However, we have tested with thicker photoresist films as well, so a discussion of this is added into section 2.2, line 153;

“Using this method, Zn tracks up to 20 μm thick can be grown, beyond which delamination from the Al substrate occurs due to internal stress. The aspect ratio is mainly determined by the photoresist thickness because electroplating fills gaps from the bottom up, replicating the photoresist profile until the Zn exceeds its thickness. Beyond this point, Zn overflows laterally, forming a mushroom-like profile with $\sim 2\ \mu\text{m}$ widening per 1 μm excess thickness. Thus, the Zn layer must remain thinner than the photoresist. In our case, the AZ4562 photoresist was $5.3 \pm 0.3\ \mu\text{m}$ thick (measured by optical profilometry), limiting Zn thickness to 5 μm and yielding a 1:1 aspect ratio for 5 μm lines. Higher aspect ratios or thicker Zn tracks will require a thicker photoresist.”

13. Section 3: Results.

Lines 167-168: Major advantage of our method over other additive processes is almost independent of the area... This argument is not reasonable, in a roll to roll process few meters/seconds can be processed, how would you proceed with the given technique to pattern 30 meters, 1 km of web? So scaling up the method becomes non competitive (without talking about all the extra steps such as lithography, etc).

We accept that other printing processes such as screen printing can be scaled more easily, and hence our statement could be misleading. Therefore, we have removed this.

Our plan for scaling to larger ultra large areas would be to employ a step-and-repeat process of $\sim 50 \times 100\ \text{cm}$ at a time, which is already used widely in the electronics industry for mass manufacture by companies such as PCBway to mass produce square metre-sized PCBs on a large scale for very low cost (see their explanatory video here - <https://www.youtube.com/watch?v=Ra79-fZpkQ4&list=PLzxd3n97eVsf63qILcg49nKTcADPyRAJ&index=4>).

It is worth stating that our method could reduce costs even further by removing the need for any subtractive etching steps from the process.

To resolve this confusion, we have removed the statement about the process being “independent of area” and have replaced it with a statement at line 203 regarding the scalability of our process as follows:

“Another major advantage of the growth-and-transfer process is that it can be easily scaled to large areas. Figure S2 demonstrates a pattern of 15 x 30 cm being demonstrated, but the only limit on scaling is in the illumination area that can be achieved in the photolithography step. For mass production on an industrial scale, the photolithography, electroplating, and transferring steps could be done in a step-and-repeat process, and the viability of this is evidenced by the fact that commercial PCB manufacturers already employ similar methods on the square metre scale¹¹.”

14. Figures 3 b, c, d, are not readable, too small figures characters, etc. My recommendation would be to put them below Figure a) on the same row, next to each other, to align them horizontally, not vertically.

We have updated figure 3 as suggested.

15. Lines 214-215: The way the results are presented requires improvement so we can really understand how reproducible is the process and the conductivity value claimed. There should be a deviation of the thickness measurement, the number of samples measured given, resistance + thickness, and this number should be at minimum 5 (from same and also for 2 other batches of samples fabricated for comparison). And finally this must lead to an average conductivity value with a standard deviation.

To further confirm this data, we repeated this measurement by making a batch of $n=7$ 2x2 cm area samples under the same plating conditions. We have updated the text in the article, and the surface profile in figure S10 to include this.

The paragraph at line 249 now reads:

“To quantify the conductivity achieved, 7 samples of 2x2 cm area were created on a UV resin substrate with a plating current density of 20 mA/cm^2 for 15 minutes. Optical profilometry data showed that this resulted in a film thickness of $10.0 \pm 0.2 \text{ }\mu\text{m}$, and the average resultant sheet resistance was $5.5 \pm 0.2 \text{ m}\Omega/\text{sq}$. This corresponds to a conductivity value of $1.8 \pm 0.1 \times 10^7 \text{ S/m}$, which closely matches the literature value for the bulk conductivity of Zn ($1.7 \times 10^7 \text{ S/m}$) and is an order of magnitude higher than most previous reports of degradable conductive materials for PCBs or in printed electronics. These are included for comparison in figure 3 (b) and (c).”

Figure S10 now reads:

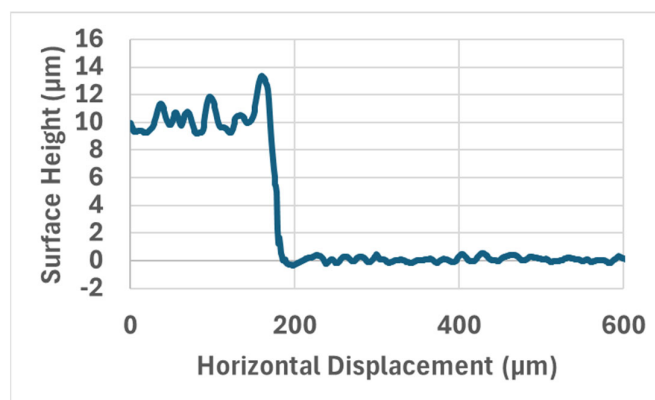


Figure S10– Representative Optical Profilometry of a step in Zn grown at a current density of 20 mA/cm^2 for 30 minutes on an Al carrier (prior to transferring to new substrate). An average thickness of $10.0 \pm 0.2 \text{ }\mu\text{m}$ was calculated from the average step height over $n=7$ samples.

16. And important, in the supplementary document, the summary table, the conductivity number cannot be rounded to be $2.0 \times 10^7 \text{ S/m}$, a value even higher than bulk value, there is should be the average number value + standard deviation for $n=5$ at least.

We have updated the value and sample number to $1.8 \pm 0.1 \times 10^7 \text{ S/m}$, as suggested above.

17. Line 244: 1000 bending cycles at 9 mm of radius. What does happen at 8 mm, it does not reach 1000 cycles?

For this test, we followed IEC 62899-202:2020, which covers dynamic bending test method for printed conductive layers. The 9 mm bend radius and 1000 cycles are within the range used for bend test reliability evaluation in this standard. And we monitored the before, during and after testing and followed the pass/fail thresholds which are stated in the standards. We have added the following text to the SI.

“These test conditions confirms that our approach complies with the IEC 62899-202:2020 testing standard for dynamic bend testing. Further testing could be done to test at extremes to gain a better understanding of failure modes.”

18.

Lines 252-253: the measured resistance in comparable, quantitative values would be appreciated, how much difference in percentage, how many samples measured. Aging tests involves statistic, a minimum of samples should be tested, an average value, standard deviation and comparison then provided.

There were 3 samples measured. We have updated the text at line 495 accordingly to include quantitative values, n values, and standard deviations as follows:

“...showing no significant change in sheet resistance even after a year of storage under ambient conditions, (n=3 samples, $14 \pm 2 \text{ m}\Omega/\text{sq}$ when fresh, $13 \pm 1 \text{ m}\Omega/\text{sq}$ when measured on day 505).”

19. Figure 4 b, c, small, and hard to read as well.

As stated at the start, figure 4 has been split into several sub-figures, but the text has now been updated as suggested.

20. pH should be written with a small p, not PH, to review all over the text.

We have made this change throughout the document.

21. Section 4: Implementation of real-world applications. Section 4.1: Touch/Sweat sensors.

There is no sweat sensor reported, 2 touch sensors are presented, basic demos. Title has to be changed, formulations in the text and discussion with references to sweat sensors, biosensors, etc, should be removed, notably lines 321-322. There is no result showing a biosensor, and no results showing that the technology can be used as biosensors, wearables, etc. The 2 touch sensors show that 2 transducing principles are applied, but at the end, these are nothing more than two zinc electrodes used to measure a capacitor and a resistor.

We have removed any references to calling the devices a sweat sensor or biosensor and instead describe it as a resistive tactile sensor. The paragraph at line 265 now reads as follows:

“As a simple demonstration of this, we produced a batch of $50 \mu\text{m}$ linewidth IDEs with 1 cm^2 area on PCL substrates, and show how these could act as an array of fully biodegradable resistive tactile sensors.

The working principle of these tactile sensors is to exploit the inherent conductivity of the salts, oils, and impurities on human fingers. When a bare human finger is placed on the active area, a conductive bridge between the fingers of the IDE is made. The impedance of this bridge is likely to be highly dependent on frequency, but the high surface area and small separation between fingers allows a large amount of current to flow when operated in DC mode”

In addition, we have adjusted the discussion on lines 279-282 to remove reference to biosensors and wearables. We instead reference other literature applications of IDEs, such as EGFETs and

photodetectors, as potential other applications of these devices for future works. The statement now reads as follows:

“In addition to this demonstrated use for human interface controls, the high surface area and short channel lengths enabled by these IDEs will have a range of other applications in areas such as biodegradable Electrolyte Gated FETs [37] or photodetectors[38].”

22. Capacitive touch sensor: polymers, etc are humidity sensitive, what was the measurement during the measurement, and what will happen to the capacitance value at different relative humidity?

This issue was also raised by Reviewer 3, and so in light of the concerns about variability of the sensor with humidity we have removed this claim from the paper. We believe that the demonstration of heaters, temperature sensors, resistive touch sensors, as well as the printed circuit boards remains sufficient evidence of real-world applications.

24. Lines 331-333: What was the power required to reach temperatures above 100°C, how stable was the heater in terms of drift of the resistance over time? How many samples tested, TCR value error, deviation for n=5 samples?

We produced a fresh batch of n=5 samples, and in all of them a voltage of 18 V was applied to achieve the temperatures exceeding 100 °C. This corresponded to a power dissipation of 4 W, and we have added a note of this in the caption of figure 7, where a graph of temperature and current vs voltage for a representative device is shown. In addition, we have added a graph to the SI showing the drift in resistance and temperature vs time to for one of these heaters when held 18 V for one hour. As can be seen, the variation over time is less than 10 %. We have updated the text at line 378 to read as follows:

“To prove the suitability of our method for this application, we produced a batch of 5 2x6 cm strip heater patterns with a 450 μm linewidth on paper substrates as shown in figure 5 (f). These devices had a resistance of $45 \pm 3 \Omega$ at room temperature, and were able to reach a temperature exceeding 100 °C when driven at 18 V. This corresponded to a heating power of 4 W, and a graph showing the average current and temperature vs driving voltage is shown in figure 5. Further thermal images of a representative sample at various voltages are also included in figure S19, and we also provide further evidence on the stability of these devices by running it continually at 18 V for one hour. It can be seen on figure S20 that there is only a ~6% drift over an hour of operation.”

In addition, we have repeated the TCR measurement on the batch of 5 samples to create an n=5 dataset, and we have updated figure 5 accordingly. From this dataset, we obtain a TCR value of $0.0061 \pm 0.04 \text{ K}^{-1}$. We have added discussion on this to line 386 as follows:

“In addition to being effective heaters, this design can also act as a temperature sensor by exploiting Zn’s linear change in conductivity with temperature. Figure 5(h) shows how the measured resistance of our 5 devices varies with temperature as they are heated up to 90 °C. It can be seen that all devices follow a linear trend that could be easily extrapolated as a temperature sensor. The slope corresponds to an average thermal coefficient of resistance (TCR) value of $0.0061 \pm 0.0004 \text{ K}^{-1}$, which is notably larger than the literature Zn value of 0.0038, and we hypothesise that this is due to added strains from the substrate causing a faster change than expected. These results show that our method can produce highly reliable devices that follow predictable and repeatable behaviour even when placed under relatively stressful conditions up to 100 °C”.

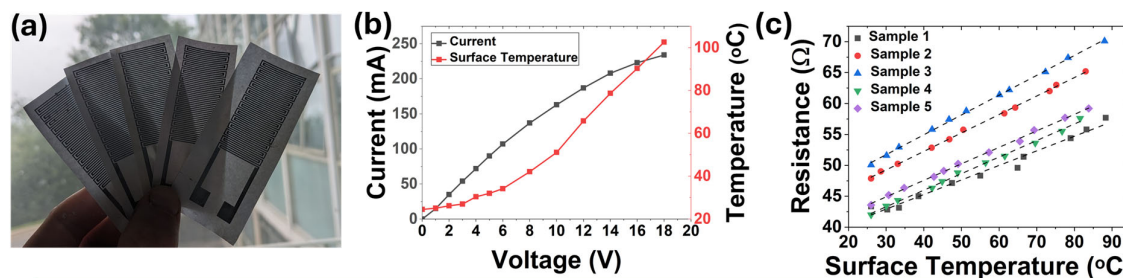


Figure 7 - (a) A batch of 5 degradable strip heater circuits produced with Zn on paper substrates. (b) The current and temperature of a heater circuit when being run at voltages from 0-18 V. This corresponds to a peak power of 4 W. (c) The resistance of the 5 circuits when being used as a temperature sensor at different temperatures – a consistent linear trend is observed.

25. Section 4.3 “Fully degradable” Microelectronics...

Again quote “” is used, so what does mean exactly Fully degradable? Is it or not fully degradable, it is not proven in any case.

We have adjusted the text; we have calculated that 99.7% of the mass of PCB is either biodegradable (in the case of paper and chitosan) or will degrade into harmless end products (in the case of Zn and carbon). In addition, the bare dies and LEDs can be recovered and reused, as we demonstrate. The calculation for estimating degradability is discussed further in the next comment.

26. What does degradable mean here ? By which means, in soil, compost? Here it is dissolved. There is no recovery of zinc, or substrate material, no clear therefore what no waste means, zinc, there is also silver, etc. where do they go? There is no proof that the zinc, silicon can degradable in soil or be compostable. Reference 37, silicon is bioresorbable, degradation in soil or compost needs to be proven. All this discussion should be reviewed and reformulated since speculative.

Zn belongs to a family of ‘degradable metals’ (alongside magnesium, molybdenum and iron), offering intermediate corrosion rates. There have been several reviews that confirm Zn’s biocompatibility, and its appropriate degradation into naturally occurring materials such as zinc oxide, zinc carbonate, or zinc phosphate in water, soil or the human body (it is bioresorbable as well). Our definition for degradability is if a material “has the ability to physically or chemically break down into non-toxic, environmentally benign end products under specific environmental conditions (e.g., moisture, heat, light, or microbial action).”

Example references are shown here;

<https://doi.org/10.1016/j.mser.2014.01.001>

<https://doi.org/10.1016/j.jallcom.2024.175313>

<https://pubs.acs.org/doi/10.1021/acsbiomaterials.4c00210>

To clarify this, we have added these references to the discussion surrounding why Zn is the most desirable degradable conductor as discussed earlier in reviewer 1 comment 3.

In the case of the silicon bare die; this is mostly made of Si which degrades to monosilicic acid in soil/aquatic environments; which is also a naturally occurring non-toxic soil compound. Admittedly this takes much longer to degrade than Zn or the substrate, but given it’s mass is <0.3% of the total PCB weight, this is not a significant concern, in our view.

As for the silver, the content is incredibly small as we are using an ultra-precise deposition system, so only 0.05 mg of silver is present on each circuit board, representing 0.007% of the total weight. Assuming a soil volume of 100g, this would put the resultant silver concentration as 0.5 ppm, which is well within the WHO guidelines on silver disposal in soil (see Nunes et al: Review of Sewage Sludge as a Soil Amendment in Relation to Current International Guidelines: A Heavy Metal Perspective. *Sustainability* **2021**, *13*, 2317. <https://doi.org/10.3390/su13042317>)

We are not intending to recover the Zn or the substrate due to their low material value and the fact that they can be disposed of naturally into the environment, this reducing the e-Waste problem if this was deployed by industry. However if full recycling was desired then the paper material could be filtered off and re-pulped, while the Zn can be recovered from the acetic acid solution via an electrowinning process similar to the one shown by Amiri et al: Zinc Electrodeposition in Acetate-based Water-in-Salt Electrolyte: Experimental and Theoretical Studies, *Chemelectrochem*, **2021**, 8,2737, <https://doi.org/10.1002/celec.20210054>.

To satisfy the reviewer, we have reformulated the discussion surrounding the degradability (line 314) to be less speculative as follows:

“In this model, the main non-degradable materials are in the LED, the Si die or the Ag wirebonds. Considering the Ag interconnects with a diameter of 30µm and an upper limit for the wire length of 1.5 mm, this corresponds to a total Ag mass of 50 µg, while the Si die and the LED both weigh around 1 mg each. The remaining mass of the PCB is made up of paper, chitosan, and 14 mg Zn (assuming 4 cm² track area with 5 µm thickness), which are all known to be degradable as discussed previously. Given the total mass of the PCB is 0.7 g, this means that ~99.7% of the device is biodegradable. The degradability of the bare Si die is currently the subject of debate, as thin (~10 µm) or mesoporous Si wafers are known to easily degrade into benign silicic acids in aqueous environments^{43,44}, but the degradability of thicker (~300 µm) wafers has yet to be proven. In the future, this device could be made to be almost entirely degradable by using a biodegradable organic LED, and by using a pre-thinned Si die to enable easy dissolution into silicic acid. In this case, a full sample degraded in 100 g of soil would reach an end concentration of 0.5 ppm Ag and 140 ppm Zn, which is within the WHO and EU limits for composting of 3 ppm Ag and 300 ppm Zn.

27. Section 5: LCA. The paper claimed notably that the method is better than printing zinc in terms of conductivity, linewidth, stability, etc. Then it would be of high interest to have the LCA between the method reported and printed zinc, especially due to the fact that the method proposed involves more steps, substrate formation, transfer, some chemicals, lithography, etc.

While the printed Zn does produce similar (albeit lower performance) devices to our work, it is inaccurate to state that the printing process has fewer steps. Our process requires 9 steps (photoresist deposition, drying, exposure, develop/seed, rinse, electroplate, rinse, transfer), and results in high performance patterns. For comparison, the work by Majee et al requires 20 printing cycles (10 Zn layers and 10 acetic acid curing layers, plus a drying cycle in between each print) to achieve their best sheet resistance of 100 mΩ/sq., which is far higher than we achieved. The work by Briand et al requires only one Zn printing step, but still requires a laser cutting step for their single-use stencil, and then 5 washes in acetic acid with a drying cycle under inert atmosphere between each wash, followed by a photonic sintering step.

In our view it is unreasonable to undertake an LCA of other groups work because 1) the source of the Zn nanoparticles and other materials used in their work is not given, 2) we have no knowledge of the energy consumption of their process steps, 3) we have no knowledge of the waste created by their process and if this waste are recycled or how this are treated at end of life. Hence, we are not able to provide a fair comparison between the processes due to insufficient information. Finally it is worth

stressing that the performance achieved is far lower than our work, which questions whether this could be used in the same applications that our work would be able to.

We have included the following brief discussion to section 5 line 428 to note this:

“We have not included a comparison of the LCA with PCBs made with printed Zn shown in previous reports as more details on material sourcing, process steps and waste generation are needed. In addition, they are not currently able to achieve performance as reported in these results. Hence, further studies are needed to provide a fair comparison between these methods.”

28. Section Methods: Materials (section should be created)

Nothing is said about the substrates materials, a section should be added on the substrates, materials used linked to suppliers. It is not clear how PHBV actually is applied, pellets from which supplier, etc, same thing for the other materials. More details should be given so the readers can reproduce the procedures.

We have added a materials section as requested. It now reads:

Line 554: “Chitosan (high Mw – CAS 9012-76-4), Glucose (CAS50-99-7), PCL (Mn~80,000, UNSPC 12162002), and Cellulose Acetate (CAS 9004-35-7) were purchased from Sigma Aldrich. ZnO, KOH, NaOH, and Acetic Acid were all purchased from Sigma Aldrich. The UV resin was e-Sun 3D printing resin,n. While the polybutylene adipate terphthalate (PBAT), Polyhydroxybutyrate valerate (PHBV), and polylactic acid (PLA)were purchased as pellets from Shijiazhuang Tuya Technology Co. Aluminium carrier sheets were “Mirror Polished” Al sheets.”

In addition, we have clarified the “Transfer Process” part of the methods section to more clearly explain how the pellets were applied. It now reads:

Line 591: “ PHBV and other thermoplastic pellets were applied by placing the Al foil carrier on a hotplate set above the melting point of the plastic (200 °C for PHBV, 80 °C for PCL, and 60 °C for chocolate), then pouring the pellets onto the surface and allowing them to melt naturally, as shown in Figure S4. Once fully melted, even pressure was applied to flatten the plastic and allow it to flow over the tracks. A non-stick silicone spray was pre-applied to the glass board to prevent sticking. After the desired shape was achieved, the assembly was removed from the hotplate and allowed to cool to room temperature. The solidified plastic was then separated from the glass board, and the aluminium carrier was peeled off, leaving the Zn tracks embedded in the PHBV.”

29. Heater/Temperature sensor measurements. Mention the model of Thermal camera used. Tape was used to glue sample on metal block, is it a thermally conductive tape? What about the emissivity value used to measure the temperature on the heater, how was done the calibration?

We have updated the description of the thermal camera on line 628 to clarify these queries. It now reads as follows:

“A Topppdon TC005 thermal camera was used (assuming an emissivity of 0.95 and calibrated by measuring PHBV samples kept in ice or boiling water) to measure the achieved temperature, using the temperature at the centre of the sample as the data point.”

Thermally conductive Al tape was used to tie the samples down to the block. We have added this information to the methods section.

30. Supplementary information. SI13. Stress testing. Title is not appropriate, it is not a stress test of the film, it is an adhesion test.

We have updated the title of the section to address this confusion. It now reads “*SI3: Adhesion, Flexibility, and Degradation Tests*”

31. Titles of tables should be above the tables, not below.

We have adjusted this as requested.

32. Table S1: there is a \$ sign for scarcely available materials, I was not able to spot that sign in the table information.

This is a mistake on our part because none of the materials listed are classified as scarcely available. We have removed this sign.

33. Table S2 has its caption on the other page. More important, the conductivity value included needs to be reviewed and comparison with laser patterned zinc layer transfer added in the comparison.

We have adjusted the conductivity value to $1.8 \pm 0.1 \times 10^7$, as per earlier discussion, and have added a row for the work by Rahimi et al.

Reviewer #2 (Remarks to the Author):

Dear Authors, thank you for this comprehensive and interesting work. I particularly appreciate the fact that you incorporated LCA in the study instead of just claiming sustainability.

Many thanks for the comments and providing your time to assess our potential journal paper. It is greatly appreciated, and you have assisted in making the paper stronger.

While the manuscript is solid and written in clear language, some concerns need to be addressed before I can recommend this study to be published. Please consider my comments:

1. You mention the issue of e-waste and in parallel discuss the applications like smart packaging. However, those single-use devices are unlikely to be treated as electronic waste and will most probably end-up in the municipal waste stream. This gives a strong motivation for designing the device in a biodegradable way, while for most other scenarios recycling would be the most desirable EoL option.

The reviewer is absolutely correct to point out that recycling is the most desirable outcome, and degradation should only be considered for “ultra-disposable” applications where this is not feasible. This should have been more clearly highlighted in our introduction, and so we have added a point on this to line 40 as follows:

“While recycling remains the most desirable outcome, the increasing ubiquity of printed electronics for inherently single-use applications such as smart packaging means that there is a growing stream of small devices which are impractical to treat as e-waste, and hence will likely end up as municipal waste.”

2. You propose the usage of Chitosan as a substrate material as well as “adhesive” layer between the Zn electrodes and the paper substrate. Especially in capacitive applications I see some issues with that: the electrical properties of Chitosan are highly dependent on ambient humidity levels, the resistance follows an exponential curve with the largest change in the lower humidity range, as described in:

Zikulnig, J., Lengger, S., Rauter, L., Neumaier, L., Carrara, S., & Kosel, J. (2022). Sustainable printed chitosan-based humidity sensor on flexible biocompatible polymer substrate. IEEE

Sensors Letters, 6(12), 1-4.

Another recent source dealing with the electrical properties of Chitosan:

E. E. Nettey-Oppong, R. Muhammad, E. Ackah, H. Yang, A. Ali, H.-W. Jeong, S.-W. Kim, Y.-S. Seok and S. H. Choi, "Development of a Sustainable Flexible Humidity Sensor Based on *Tenebrio molitor* Larvae Biomass-Derived Chitosan," Sensors, vol. 25, no. 2, p. 575, 2025

Considering this, your application examples of the IDE sensors would not work reliably. In general, to guarantee stable operation of electronic devices, an inert substrate material should be preferred to avoid cross-talk.

This issue has also been noted by Reviewer 1, and in light of these reliability concerns we have removed the claim of capacitance-based touch sensors from the paper.

We have kept the claim of resistive tactile sensors because their working mechanism would require a much larger change in the substrate's electrical properties to cause a failure or problem to emerge in our circuit. To confirm this, we placed the circuit (seen in figure 5c) within an environmental chamber at 35C and varied relative humidity from 50% to 95% and were still able to operate the circuit.

Whilst the change in electrical properties doesn't impact our circuit, we agree this could have caused erroneous behaviour in other circuits, so we have added the following text to line 358 of the manuscript;

"It is worth noting that the electrical properties of the adhesive layer (Chitosan) are dependent on ambient humidity levels; the resistance follows an exponential decrease as relative humidity is increased⁵². Tests were conducted under varying relative humidity levels (50-95%) and the circuit remained operational. However, for more sensitive applications, humidity might interfere with sensor response or circuit performance."

And added the following reference;

52 Zikulnig, J., Lengger, S., Rauter, L., Neumaier, L., Carrara, S., & Kosel, J. (2022). Sustainable printed chitosan-based humidity sensor on flexible biocompatible polymer substrate. IEEE Sensors Letters, 6(12), 1-4.

3. For most devices, especially in the field of printed electronics, the substrate material accounts for the largest part of the mass. Historically, this was somewhat misleading for researchers to assume that the focus on the substrate reduces the environmental impact the most, while recent research has shown that silicon components as well as silver inks pose as environmental hotspots:

Zikulnig, J., Carrara, S. & Kosel, J. (2025). A life cycle assessment approach to minimize environmental impact for sustainable printed sensors. Scientific Reports. Can you please elaborate that further in the context of your work?

This is a similar conclusion from our work also. The life cycle environmental impact in many categories is higher for the silicon IC than the substrate, despite having a lower mass (see <https://doi.org/10.1038/s41598-024-79732-1>), so great minds clearly think alike!

However, the substrate still is a significant contributor to the environmental profile in many impact categories, and this has more of an impact for larger electronic circuits such as those used in TV remote controls than more compact circuits such as smart watches (as shown in our earlier paper). In addition, the substrate typically contributes the most to the overall mass of a PCB. From an e-waste perspective, this increases the volume sent to landfill, requires more landfill space, requires more handling and transportation etc. Reducing substrate mass could represent a relatively 'low-hanging

fruit' for the electronics industry to improve the industry's environmental impact. In our view, it's essential not only to improve the life cycle assessment (LCA) profile of electronic assemblies but also to minimise the total mass of waste generated.

To elaborate in the context of our work, the following section at line 460 has been added;

“Based on the LCA data, our work demonstrates several strategies for improving the environmental profile of electronic assemblies. Electronics often rely on Critical Raw Materials (CRMs) such as silver (Ag) or gold (Au) as well as Copper, which have significantly higher environmental footprints than Zn [15, 51]. Our approach also enables improved recovery of valuable components, such as ICs, which often contribute more to the overall environmental impact than the substrate or conductive tracks. Finally, the approach should enable lower EoL footprints, as recycling is made simpler and is beneficial for situations where recycling is impractical.”

4. Please describe the resist stripping process. Does it affect the Zn layer and why (not)?

The resist is stripped by dipping the sample in a bath of acetone. This does not affect the Zn layer because Zn is insoluble in acetone, and the residual adhesion of the tracks to the carrier remains sufficient to prevent them from delaminating as long as high pressure sprays are not used. We have updated the methods section on line 586 to clarify this as follows:

“After plating, the resist is stripped by dipping the sample in a bath of acetone. This does not affect the Zn layer because Zn is insoluble in acetone, and the residual adhesion of the tracks to the carrier remains sufficient to prevent them from delaminating.”

5. On page 2 you describe that degradable metals have an “awkward” melting point – I don't think this wording is appropriate, it sounds like slang.

We have adjusted the sentence to use more formal language. The sentence now reads

*“This is because its **low** melting point and tendency to crystallise into rough films with....”*

6. The IDE resistive touch sensor: It is perhaps inaccurate to say that skin is conductive because of sweat – it is rather because of salts and impurities present in the skin and the impedance is highly dependent on the frequency. Please correct that.

This was also raised by reviewer 1 and we have adjusted the discussion accordingly. Line 268 now reads:

“The working principle of these tactile sensors is to exploit the inherent conductivity of the salts, oils, and impurities on human fingers. When a bare human finger is placed on the active area, a conductive bridge between the fingers of the IDE is made.”

7. For temperature sensing, you mention a change in resistance of 0.7 Ohm/degC. Please express that in terms of TCR and compare with Zn bulk metal.

This issue was also raised by reviewer 1, and we have since repeated this measurement on n=5 samples on paper substrates. All samples showed a similar slope, and the average TCR measured was 0.0061 ± 0.04 , which differs substantially from the literature value (0.0038). We hypothesise that this is due to other interactions from the thermal expansion of the substrate, and we have updated the figures and added discussion on this to line 386 as follows:

“In addition to being effective heaters, this design can also act as a temperature sensor by exploiting Zn's linear change in conductivity with temperature. Figure 5(h) shows how the measured resistance

of our 5 devices varies with temperature as they are heated up to 90 °C. It can be seen that all devices follow a linear trend that could be easily extrapolated as a temperature sensor. The slope corresponds to an average thermal coefficient of resistance (TCR) value of $0.0061 \pm 0.0004 \text{ K}^{-1}$, which is notably larger than the literature Zn value of 0.0038, and we hypothesise that this is due to added strains from the substrate causing a faster change than expected. These results show that our method can produce highly reliable devices that follow predictable and repeatable behaviour even when placed under relatively stressful conditions up to 100 °C"

8. Thermal imaging: Please provide information on the thermal emissivity (epsilon) used for temperature measurement.

This issue was also raised by reviewer 1. The emissivity was assumed to be 0.95, and we have adjusted the methods section line 628 accordingly

"A Toppdon TC005 thermal camera was used (assuming an emissivity of 0.95 and calibrated by measuring PHBV samples kept in ice or boiling water) to measure the achieved temperature, using the temperature at the centre of the sample as the data point."

9. Why are you using wirebonding? Flip-chip bonding could reduce the material needed for chip integration and with the high resolution of your method, replicating the chip footprint should not be an issue. Please also briefly discuss the issues of thermal processing with temperature sensitive substrate materials, like PLA, Chitosan, Cellulose Acetate.

The reviewer also correctly points out that many of the materials used are unsuitable for high temperature processing, and deform even at modest heating of 80-100 °C. It is for this reason that a paper/chitosan substrate was used, as we found that this showed minimal deformation even up to 150 °C. It is also for this reason that we were unable to successfully achieve flip-chip bonding. While our pattern resolution is more than sufficient to match the chip footprint, we found that none of our substrates could withstand the >200 °C temperatures required to melt the SAC solder used in flip-chip bonding, and so we used wire bonding instead due to its lower temperature constraints. It is also worth stressing that less material is used in this wire bonding approach than using solder balls or conductive silver adhesive.

In addition, the XTPL wirebonding approach allows for thinned ICs to be used in the future, which would degrade much quicker in soil/water than bare dies, which opens up a possibly more another generation of transient electronics.

We have added a discussion on this to section 4.3, line 297 as follows:

"For this, paper was used as the biodegradable substrate, with a thin layer of chitosan to facilitate Zn transfer. This substrate was selected due to its minimal deformation when annealed at temperatures up to 150 °C, which is the minimum required to achieve good conduction from UPD silver wire bonds. In contrast, bioplastics such as PHBV, PLA, and cellulose acetate were unsuitable, as they experienced significant deformation even at moderate processing temperatures (around 80 °C), while PCL deformed above 65 °C. These thermal limitations further underscore the advantage of the G-T method, as other printing techniques that require annealing would not permit the deposition of conductive tracks without causing unacceptable substrate distortion. Additionally, this poses challenges for component soldering and processes such as Flip-Chip bonding; however, the adoption of lower-temperature solders could help mitigate these issues in future developments."

10. I have an issue with the EoL of the biodegradable sensor. How can the GWP be negative, when it degrades? Usually, under biodegradation, the CO₂ stored in the material is released. Please elaborate on this further!

A negative GWP in an LCA in this case indicates that the PCB substrate removes more greenhouse gases (GHGs) from the atmosphere than it emits over its life cycle. Biodegradable materials like paper, PLA, PCL, or cellulose-based substrates are often made from biomass, which absorbs CO₂ from the atmosphere during growth (ie. via photosynthesis). If this carbon stays stored in the material or is biodegraded in a way that does not release all of it as CO₂ or CH₄, the net carbon impact can be negative. In addition, carbon retention in compost or soil, particularly if the material ends up improving soil organic carbon. That carbon is effectively sequestered, offsetting other emissions. It is difficult to know from the LCA data which of the two effects dominates, so we have added this text to line 455:

“The GWP is negative due to two reasons; firstly, the biodegradable substrate material absorbs CO₂ from the atmosphere during growth (due to photosynthesis). As much of this carbon is known to biodegrade in a way that does not release all of it as CO₂ or CH₄, the net carbon impact can be negative. Carbon retention in soil can improve soil organic carbon, so this is also sequestered, offsetting other emissions.”

11. FR-4 substrates contain flame retardants. How would you mitigate the issue of flammability with your devices?

Magnesium hydroxide is a common flame retardant used in commercial fabrics that is both biocompatible and low environmental impact. Future work on improving flame retardancy would involve dispersing Mg(OH)₂ in safe solvents to make a flame-retardant spray that could be applied to the paper substrate. We have added discussion on this to line 362 as follows:

“In the event of commercial application, these PCBs could be made to comply with flame retardancy regulations by incorporating magnesium hydroxide (Mg(OH)₂) into the paper matrix, or by applying it as a dispersion in a spray post production. Mg(OH)₂ is a biocompatible, non-toxic, and environmentally friendly flame retardant that is already used in commercial products⁵³.”

Reviewer #3 (Remarks to the Author):

The manuscript presents an overmould printing method for fabricating fully degradable electronic circuits using bulk zinc tracks embedded into biodegradable substrates. This approach enables high-resolution patterning, near-bulk conductivity, and compatibility with a wide range of substrate materials. The authors demonstrate various devices including touch sensors, heaters, and microelectronic circuits, and provide a life-cycle analysis showing reduced environmental impact compared to traditional FR4 PCBs. The method is promising for sustainable electronics and low-cost biodegradable IoT systems. However, several issues should be addressed before the manuscript can be considered for publication:

1. The manuscript lacks quantitative data on fabrication yield and reproducibility across substrates and pattern geometries. It is unclear how robust or scalable the process is in practice.

Similar queries about reproducibility have been raised by the other reviewers, and to this end we have demonstrated the repeatability of our method over different substrates and devices by producing n=5 batches of most of the test devices, which include paper, PHBV, and PCL as substrate materials. We found that devices with feature sizes greater than 100 μm showed close to 100 % yield, and the key factor which affected the reproducibility of devices with smaller feature sizes was parasitic deposition outside of the desired area. This deposition would occur at defect points in the photoresist template

caused by dust particles during resist deposition, or resist peeling from the H₂ bubbles created during the seed layer deposition. These would cause shorts between lines and thus reduced yield. To quantitatively demonstrate this, we produced a large batch of interdigitated electrode devices (IDEs) with linewidths down to 2.5 μm . IDEs are extremely sensitive to shorting defects, and so even a single defect will cause a short in the device. We produced 8 batches, containing a total of 516 devices (86 of each linewidth), and show a yield curve of unshorted devices vs linewidth, which we have added to the SI. As can be seen, yield drops rapidly for feature sizes less than 25 μm , but this could conceivably be improved in the future by performing the fabrication in a cleanroom environment.

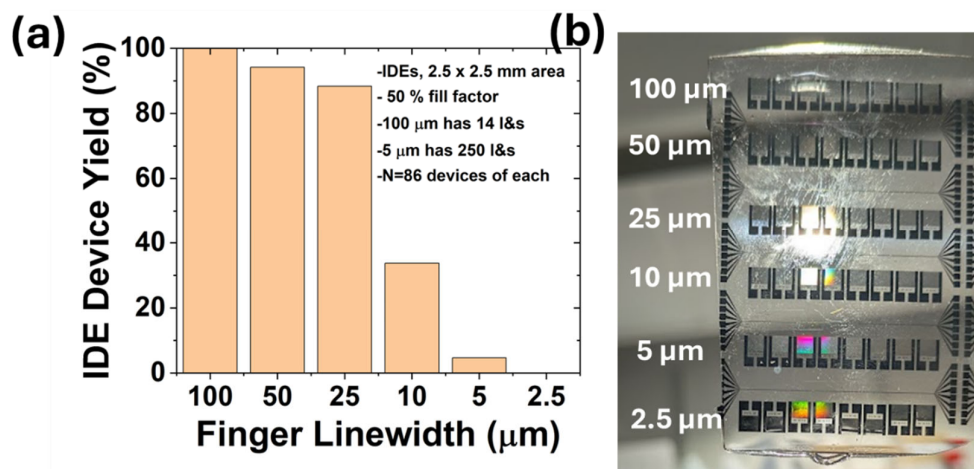


Figure S24 – Yield tests performed using interdigitated electrode patterns with different linewidths on UV resin substrates. The devices have a 2.5x2.5 mm active area, consisting of line and space patterns with 50 % fill factor, and were fabricated in batches as shown in (b). Note the diffraction patterns occurring on the finer linewidth samples. These were fabricated outside of a cleanroom, meaning the yield drops rapidly at linewidths below 25 μm , but cleanroom processing and further optimisation could improve this.

In addition, we have added the following to line 193 to discuss this:

“To investigate the reproducibility of these fine feature sizes, we produced 8 batches of IDEs with 2.5 x 2.5 mm area with different linewidths from 100 μm to 2.5 μm , totalling 86 devices for each linewidth. IDEs are highly sensitive to defects, as any parasitic deposition will cause an instant short circuit between the contacts, which makes it for an ideal testing platform for our process yield. As can be seen in figure S23, the yield of unshorted devices remains above 85 % for features as small as 25 μm , but falls off rapidly as the finer pitches can be shorted by ever smaller defects. It should be noted that all fabrication was done outside of a cleanroom, and so production in a more controlled, dust-free environment would likely improve these results substantially.”

2. The long-term operational stability of the devices is not sufficiently evaluated. While bending and humidity resistance are shown, extended testing under realistic conditions is needed to assess reliability.

The stability of our samples under ambient conditions is evidenced by the data in figure 4, which shows negligible change over more than a year of storage. As further evidence for the stability of our devices under real operating conditions, we performed a test of n=5 devices carrying a high current load (150 mA in a 2 mm track) at elevated temperature (80 $^{\circ}\text{C}$) at 50 % relative humidity. While a slow drift in resistance is observed, it amounts to less than 0.3 % change per day. This data has been

added to the SI as shown below. In addition, we have added the following line to the discussion at line 497:

“ This high stability was also observed when unencapsulated devices were forced to carry large current loads at elevated temperatures for weeks at a time, as shown in figure S22, which demonstrates that they should have sufficient reliability for consumer use”

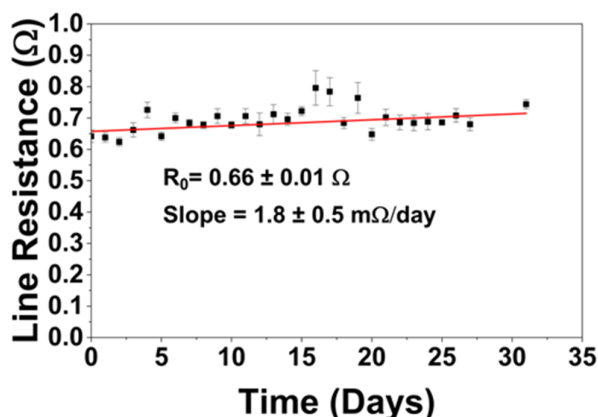


Figure S22 – Line resistance of $n=5$ Zn devices, consisting of 60 mm length lines with 2 mm width on a paper substrates, as they are stressed by constantly carrying a current of 150 mA while held at elevated temperature (80 °C) under atmospheric conditions (~50 % RH).

It is worth stressing that reliability could be significantly improved with encapsulation. Our group does a lot of work on reliability and a full reliability test (thermal cycling, flame retardancy, Voltage switching, damp heat) seems out of the scope of this paper. This paper is primarily to demonstrate the new and novel approach for PCB fabrication and provide some confidence in the approach by demonstrating circuits and some basic reliability tests. The paper is already quite long and to add an extra 5-10 pages on a comprehensive reliability test seems unnecessary.

3. The comparison with commercial copper-PCBs is overstated. Direct benchmarking of full circuit performance, including thermal limits, signal integrity, or integration density, is absent.

We have added a discussion comparing the limitations of our method to conventional PCBs (see our response to point 5 below), and have also included a table comparing the key performance indicators you requested

Table S4 – A comparison of literature key performance indicators for PCBs made from conventional materials and from biodegradable materials in our method.

PCB Material	Maximum Operating Temperature (oC)	Minimum Routing Pitch (μm)	Dielectric Loss Tangent	Dielectric Constant
FR4[34]	~150 oC	100 (commercial), 50 (HDI/LDI)	0.026 (2 GHz)	4.4 (2 GHz)

Polyimide[35]	>260 oC	100 (commercial), 25 (Fine flex)	0.015 (1 GHz)	3.24 (1 GHz)
Paper[36]	~120 oC	10 (our work)	0.09 (5 GHz)	3.2 (5 GHz)
PHBV[36]	~80 oC	10 (our work)	0.008 (5 GHz)	2.9 (5 GHz)
Chitosan[37,38]	~120 oC	10 (our work)	0.32 (1 MHz)	3.0 (1 MHz)

4. Recommend citing ACS Nano 2021, 15, 9328; Nat. Commun. 2022, 13, 3223; TrAC, Trends Anal. Chem. 2021, 143, 116369; and TrAC, Trends Anal. Chem. 2018, 102, 236 in the Introduction to better contextualize recent developments in degradable microelectronics and biosensors.

We thank the reviewer for their suggestion. There are papers already included summarising recent developments.

5. Known limitations of the method (e.g., thermal sensitivity of substrates, Zn oxidation, adhesion failures on certain materials) are not discussed, which hinders realistic evaluation of the technology's scope.

We have added a new section (4.5) to line 397, where we discuss the limitations of the method. The new section now reads as follows:

“The above devices are designed for specific disposable applications, where the priority is minimising environmental impact. While we have demonstrated circuit performance under a range of environmental conditions, it is important to discuss the limitations imposed by degradable materials to clarify where their use is most appropriate.

The most significant limitation of these devices is their thermal performance. Although paper-based samples can withstand temperatures up to 150 °C for short periods, all the biodegradable plastics tested here have low glass transition temperatures. As a result, some of them deform at temperatures above 80 °C. Additionally, the presence of a native oxide layer on the surface of the Zn tracks makes soldering components challenging. Therefore, new surface-mounting approaches are needed, such as surface finishing of solder points (as in conventional electronics) or the use of novel conductive adhesives. The high-frequency performance of these substrate materials may also be lower than that of conventional PCBs based on FR4 or polyimide, primarily due to water ingress into the substrate. However, certain biopolymers, such as PHBV, have previously been shown to match or even outperform conventional substrate materials in this respect. A comparison of these key parameters is provided in Table S4. Finally, the fabrication method requires casting a continuous liquid film for effective transfer. Consequently, it cannot be directly applied to non-meltable materials such as glass, and integration with porous substrates like TexWipes results in non-breathable films with poor adhesion. Despite these limitations, the intended applications (such as smart packaging, IoT devices, disposable test kits, and environmental sensors) remain unaffected, highlighting the strong potential for future use.”

6. Key datasets such as sheet resistance, temperature response, and degradation time are presented without error bars or information on measurement variability. Statistical analysis is needed to validate claims.

This important issue was also raised by reviewer 1, and so we have done repeat measurements where necessary to provide at least n=5 datasets for sheet the sheet resistance, adhesion strength, thickness measurements (resulting in an estimated conductivity), temperature response, TCR value, heating

efficiency, and n=3 samples for sheet resistance vs degradation time. The specifics are discussed in earlier responses.

7. The recycling demonstration is qualitative. Quantitative data on component recovery efficiency, functional performance after reuse, and degradation of parts during disassembly would greatly strengthen this part.

We have performed the recovery process on a batch of n=10 of all the critical components in the circuit (LEDs, resistors, and microcontrollers) and added quantitative data on their performance metrics (resistance and forward bias voltage) to the SI to show how they are affected by the process as shown below. A quantitative metric for the performance of the microcontrollers is harder to define, but we can confirm that all 10 of them work, and are still able to interface with an external computer and be reprogrammed after recovery. In addition, we have updated the text in section 4.4 line 347 as follows:

“To prove that the components can survive this process, we performed the recovery process on n=10 samples of 220 Ω resistors, LEDs, and microcontrollers, and tested their performance before and after. We found that all microcontrollers remained fully functional after recovery, while the LEDs showed no significant change in forward bias voltage. The resistors were found to exhibit a small (0.5 %) rise in resistance over the process, but this remains well within the ± 5 % design tolerance of the components. These results show that, in addition to a linear lifetime based on degradation at the end of usefulness, our method can also enable a circular life cycle where valuable components can be easily re-used⁵¹ while the low-cost substrate is safely disposed of. This proves that our work can cater to a wide range of use cases, covering mid-tier devices where components still have some value at the EoL to “ultra-disposable” applications where the best solution is to simply let the device degrade.”

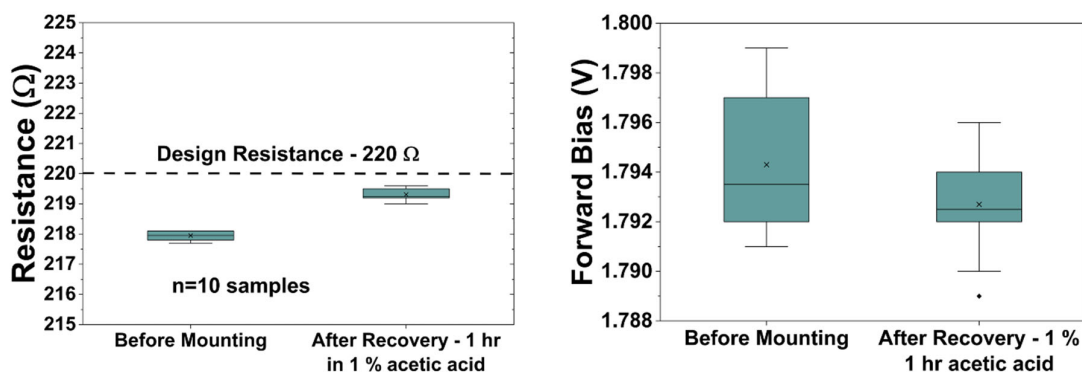


Figure S23 – Performance metrics of resistors and LEDs before and after being put through the recycling process described in section 4.4. As can be seen, there is a negligible change in forward bias for the LEDs, and the change in resistance is well within the 5 % design tolerance of the 220 Ω resistors.

Reviewer 1

The authors made large efforts to improve significantly the paper. There are still a few points to be addressed before acceptance for publication.

Firstly, we have found Reviewer no. 1 comments very useful. Clearly you are an expert in this area, and we very much respect your feedback and welcome your constructive criticism.

Comment 1

Biodegradation of Zinc PCB. I think the numbers and process to reach the conclusion of 99% biodegradable can be questioned according on how you consider what is safe, not safe, degrades or compost. Because of this I would:

- Remove 99% in the title. It can appear elsewhere, but this is claim is incomplete, not considering all the aspects, and therefore could be debated.

We agree and have softened the wording of the title to the following:

"Designing for End-of-Life Electronics: Additive Electroplated Zinc Conductive Tracks enabling High-Performance Printed Circuit Boards with Low E-Waste Footprint"

-Saying that zinc is safe once degraded should be removed as well. Zinc is a heavy metal and limited in soil content. I believe that following the composting regulations the quantity of zinc would be too much (we did the test) and therefore the PCB would not be compostable. It is easy to say everything degrades in soil, but in practice it is not applicable. I agree that the PCB biodegrades in the conditions given, but still care should be taken about that this is safe (answer to comment 8 says that Zn safely degrades), release benign products, etc.

We understand the reviewer's concern that "safe" and "benign" are terms which require a very high standard of proof, and so we have removed any reference to these words in the text. We have addressed this further in comment no. 26. However, we have replaced "can safely degrade at their end-of-life (EoL) with:

"...and could degrade into low-toxicity end products at their end-of-life"

Comment #3:

Indeed, the comment asked to precise in the introduction the statement mentioned in the abstract, to justify it, why zinc is the most desirable degradable conductor. However, in the information added. I do not understand why emphasis is put on biocompatibility, we are talking about degradable conductors in the environment, and therefore the discussion should be not limited to the fact it is biocompatible (this means with human body and also in a limited daily quantity if bioresorbed), listing and comparing with sodium and calcium, that are not really adapted for making electronic boards (iron not ideal, but could be considered). W, Mo, Mg are missing in the discussion for instance.

This comment overlaps closely with the following comment, and so we address this more fully below.

On the latter, the reviewer is correct to point out W, Mg, Fe and Mo are alternative materials. However, when comparing their stability, Fe (in thin film form), Mg and W are far less than Zn

(Mo and Zn are similar from our tests). Zn possesses a higher conductivity and is more abundant than Mo, while also being permissible in far higher quantities. So overall, Zn is the preferred metal. See below comment (no. 28), but from lines 58, more discussion of 'transient metals' is given.'

Comment #26:

All papers listed to backup the following discussion mainly address clinical and in body applications. We have published different papers on zinc and transient electronics and we look at the literature. Everyone claims and refer to some paper that Zinc degrades well in soil, etc, but looking at all references and mechanisms, they are related to implantable, in body, in liquid degradation (Such as PBS). So what is described below and the references given are for zinc in body. What about zinc outside the body, have you found any references on studies looking at zinc degradation in soil ? We have not. In the text below mechanisms are given, etc, and soil is listed among other environments, this requires proper references if you want to claim that zinc degradation has been studied demonstrated in soil.

There are multiple papers which discuss the impact of Zn in soil or in agricultural settings (see links at the bottom), where it usually takes the form of a water soluble Zn^{2+} ion. It is indisputable that Zn is an essential element for most life forms but, like almost any material, excessive concentrations can become a pollution concern.

The references at the bottom of this comment discuss the pollution issues from industrial Zn output, but also point out that cases of Zn deficiency in soil are a far more common issue. Indeed, the pollution concerns from Zn primarily arise from the overuse of Zn as a fertiliser/additive in a similar way to how nitrate overuse is known to cause negative second-order effects. In both of these cases, the definition of whether they are "hazardous", "harmless", or "essential" products within the soil depends substantially on the context. Hence we understand the reviewer's concerns with our use of "benign" or "harmless" end products to describe degraded Zn, but we would also contend that the permissible quantities for Zn in soil remain higher than for all the other viable metals that could be used as conductors in a PCB (unless the instability issues with Fe or Mg could be resolved), and therefore the Zn is the most desirable material currently.

A full study of the final fate of Zn^{2+} ions in composting waste would be outside the scope of this paper, and so to satisfy the reviewer we have added a discussion noting the discrepancy between the studies on biocompatibility vs soil safety in electronic products. While we emphasise the need for further study and discussion, we contend that Zn remains the "least bad" of the available options, which is why we pursue it in this work. In addition, we have been careful to remove further use of the loaded terms such as "safe", "benign", "harmless", "non-hazardous" etc within the text of the paper to highlight the fact that very few materials can be unambiguously classified by these terms at all concentrations.

"Among the degradable metals, zinc (Zn) is the most frequently used because it has the best balance between abundance, biocompatibility^{26,27}, conductivity, and stability²⁸⁻³⁰. Zn has an environmental footprint ~ 100x lower than Ag³¹, and a high conductivity of 1.7×10^7 S/m . It is an essential trace element that is required by the human body in quantities almost as high as iron, and is rated as safe to use for human biological applications such as smart implants or topical wound healing creams such as SudocreamTM. Despite its proven biocompatibility, the suitability of Zn for soil applications remains more understudied. Zn degrades in soil to naturally occurring compounds such as ZnO , ZnCO_3 , or $\text{Zn}_3(\text{PO}_4)_2$ depending on the soil composition²⁵, and it is frequently used as an additive to compost or animal feed, in order to

accelerate plant growth². Conversely, excessive concentrations are also known to act as a pollutant^{3,4} in a similar form to how overuse of nitrate fertilisers is known to cause eutrophication issues⁵. Hence, permissible limits of Zn use in compost do exist, ranging from 150-800 ppm depending on legislation⁶⁻⁸. Certain metals such as Fe, Ca, and Mg have higher permissible limits than this, but they remain too unstable for use in a practical device, while more stable metals such as Cu or Mo have far lower permissible limits of 50-150 ppm and 10 ppm respectively⁹. Hence, Zn is likely to be the “least bad” metal for use in degradable electronics, but further research into this important field is clearly needed.”

References for the reviewer in regards to soil toxicity of zinc

Kaur, H., Srivastava, S., Goyal, N., & Walia, S. (2024). Behavior of zinc in soils and recent advances on strategies for ameliorating zinc phyto-toxicity. *Environmental and Experimental Botany*, 220, 105676.

Noulas, C., Tziouvalekas, M., & Karyotis, T. (2018). Zinc in soils, water and food crops. *Journal of trace elements in medicine and biology*, 49, 252-260.

Hussain, S., Khan, M., Sheikh, T. M. M., Mumtaz, M. Z., Chohan, T. A., Shamim, S., & Liu, Y. (2022). Zinc essentiality, toxicity, and its bacterial bioremediation: A comprehensive insight. *Frontiers in microbiology*, 13, 900740.

Westendorf, J. (2012). Scientific Opinion on safety and efficacy of zinc compounds (E6) as feed additive for all animal species: Zinc oxide, based on a dossier submitted by Grillo Zinkoxid GmbH/EMFEMA. *EFSA Journal*, 10(11), 2970.

Another point we have made is to calculate the mass of Zn present on the PCB and evaluate how much this could impact Zn environmental limits within soil sample.

Thresholds under Directive 86/278/EEC (relating to sewage-sludge application) cite soil limits of 150-300ppm. Using our PCB design, we calculate 0.0064g (6.4mg) of Zn is present on the PCB. When comparing to the lower Zn EU limit (150ppm), 150mg of Zn is permitted to be present per kg of soil. The average background soil levels across the EU are typically 41mg/kg.

Therefore, disposal of this PCB into 1kg of soil means we are well within the limits by a factor 3.

In fact, the Zn from each PCB used in this study can be disposed of into approximately 58.7g of soil and remain within the lower EU threshold for Zn in soils.

We are not experts in soil or environmental science, or environmental legalisation, so we feel uncomfortable placing such information into a journal paper, despite our ‘high’ confidence in the calculation. However, we have added this text, which will allow readers of the paper to make their own assessment in future, and also provides an important caveat for anyone working in this area, which hopefully will provoke more studies into this work;

“For this PCB example, Zn mass was calculated at 6.4mg, which the authors believe to be within the thresholds of Zn content defined under EU Directive 86/278/EEC (relating to sewage-sludge application), which cite soil limits of 150-300ppm. Whilst Zn is used in bioresorbable applications, its full safety in outdoor environments merits further attention. We therefore recommend additional studies to understand its degradation pathways in different media (soil, water, landfill).”

There is also an issue with reference 21 which refers to PEDOT:PSS. I believe this reference or numbering should be changed. And I would really like to have a look at this reference to see what the conclusions are and they were obtained.

We have double-checked the reference, and we believe that the link now works. We include a printscreen of the paper's publication for the reviewer's reference. We believe that the assertion of "biodegradability" in their paper centres around a cytotoxicity study.

The screenshot shows the APL Bioengineering journal website. The top navigation bar includes links for HOME, BROWSE, COLLECTIONS, PUBLISH WITH US, and ABOUT. The article is from Volume 7, Issue 1, March 2023. The title is 'Tailoring inkjet-printed PEDOT:PSS composition toward green, wearable device fabrication'. It is part of a Special Collection: 'Emerging Technologies in Wearable Sensors'. The authors listed are Marina Galliani, Laura M. Ferrari, Guenaelle Bouet, David Eglin, and Esma Ismailova. There is a 'Check for updates' button and a link to 'Author & Article Information'. The article is published in APL Bioeng. 7, 016101 (2023) with a DOI of 10.1063/5.0117278. The article history is also available. The article content is divided into sections: I. INTRODUCTION, II. RESULTS, and A. Inkjet printable PEDOT:PSS ink formulation. The abstract mentions that inkjet printing remains one of the most cost-efficient techniques for device prototyping and manufacturing, offering considerable freedom of digital design, non-contact, and additive fabrication. When developing novel wearable devices, a balanced approach is required between functional, user-safe materials and scalable manufacturing processes. Here, we propose a tailor-made ink formulation, based on non-hazardous materials, to develop green electronic devices aimed at interfacing with humans. We demonstrate that developed ink exhibits high-resolution inkjet printability, in line with theoretical prediction, on multiple wearable substrates. The ink's chemical composition ensures the pattern's enhanced electrical properties, mechanical flexibility, and stability in water. The cytocompatibility evaluations show no noxious effects from printed films in contact with human mesenchymal stem cells. Finally, we fabricated a printed wearable touch sensor on a non-woven fabric substrate, capable of tracking human steps. This is a step toward the development of green wearable electronics manufacturing, demonstrating a viable combination of materials and processes for biocompatible devices.

Comment #15:

Thank you for the new experiments made on the conductivity of the Zn films, but I have a serious issue still with the value reported which for me questions the robustness of the methodology used. How can you reach conductivities higher than the bulk zinc value ? How can you explain this ? And this value is still now referenced in the comparison table.

The literature value for the conductivity of Zn is 1.7×10^7 S/m and our results obtained a value of $1.8 \pm 0.1 \times 10^7$ S/m. Hence, our result is within margin of error of the theoretical value, and so we would argue that our measurement matches the theoretical value within statistical significance. However, we understand that the use of a headline 1.8×10^7 could cause confusion, and so we have adjusted all references to this value to say " 1.7×10^7 (within experimental error)". In addition, we have adjusted the text on page 11 to say the following:

"Because our measured value falls within the experimental error of the theoretical value, we shall henceforth refer to our conductivity as the literature value of 1.7×10^7 S/m for clarity."

Comment #24:

I have also serious concerns on the methodology used to measure the TCR coefficient (and therefore the temperature in the experiments), since the TCR value found is 50% higher than the bulk value, how can this be physically possible ?

Our response to this comment overlaps with our response to comment 29, and so they are addressed simultaneously below

Comment #29:

How was selected this emissivity 0.95, was it measured ? From literature ? Did you measure temperature on substrate or on zinc track ?

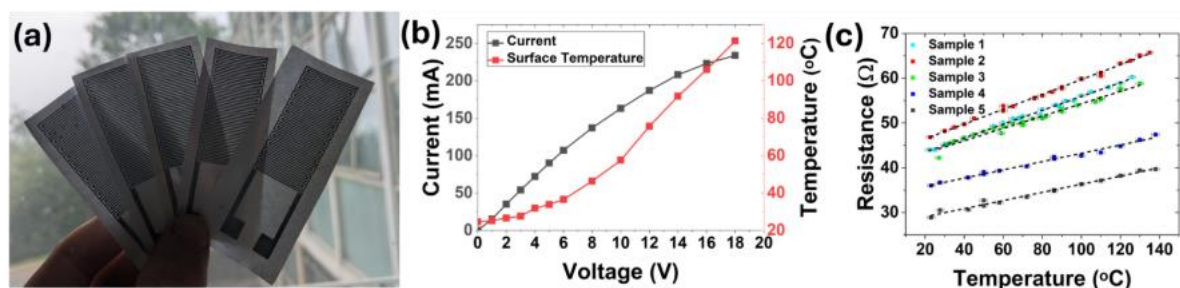
To satisfy the reviewer, we have removed experiments conducted with thermal cameras and have repeated all measurements using two separate and pre-calibrated thermocouples. The thermocouples were both taped to be in thermal contact with the same point on a given sample, and they were found to have a maximum difference in measured temperature of 3 °C, and so we applied error bars of ± 2 °C to all measurements. We have updated figure 7 and S19 accordingly, and the measured TCR value came out to 0.032 ± 0.004 , which is closer to the theoretical value of 0.037, and we hypothesise that the remaining differences may be due to CTE mismatches between the substrate and the metal.

The following changes have been made to the text;

Line 410 now reads:

“Their slope corresponds to an average thermal coefficient of resistance (TCR) value of $0.0032 \pm 0.0004 \text{ K}^{-1}$, which is differs slightly from the literature Zn value of 0.0037, and we hypothesise that this is due to interactions between the thermal expansion of the Zn and the substrate.”

Figure 7 now reads:



Line 651 now reads:

“Two calibrated thermocouples (A Hannah instruments K-type thermocouple and an OWON B35T+ thermocouple) were used to measure the sample temperature, which were both taped to the sample and ensured to be in good thermal contact. The maximum deviation between the two measurements was 3 °C when the average reading was 130 °C, and so we apply error bars of ± 2 °C to the measurement.”

Figure S19 now shows the following:

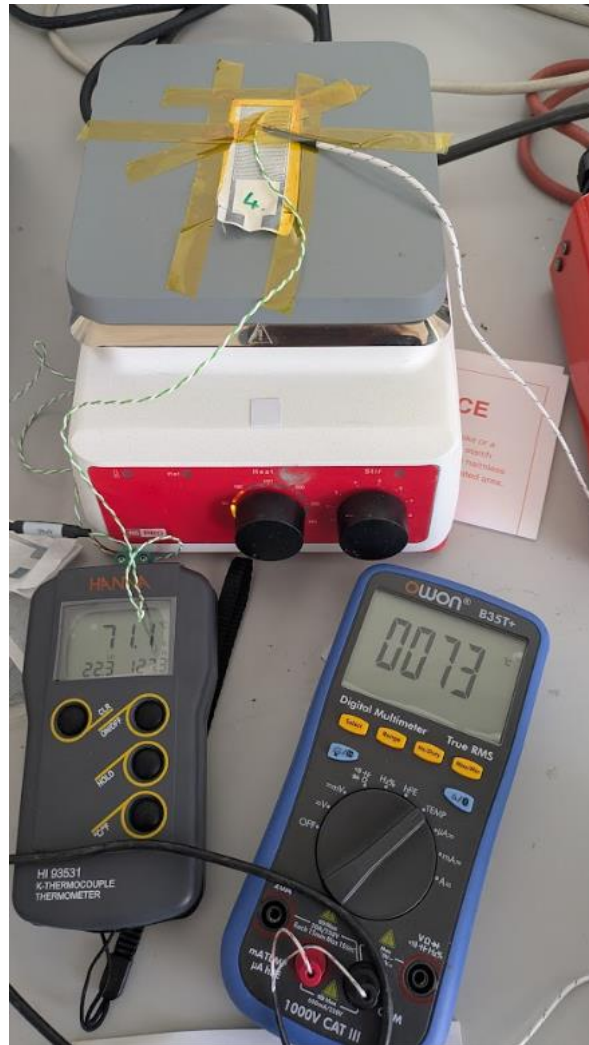


Figure S19 – The setup for measuring the TCR of the heaters/temperature sensors. The sample is fixed to the hotplate using double sided Al tape within the serpentine region in order to assist in thermal contact.

Reviewer no. 2

The authors have made a substantial effort to address the reviewers' concerns. The revised manuscript includes additional data on fabrication yield, stability, and recycling, which improves the overall presentation. However, several critical issues regarding experimental rigor and data reliability remain unresolved. In its current form, the manuscript still does not meet the standards for publication. The following points require attention:

1. Data presentation remains weak. Several key images (e.g., Figures 1f, 3a, 9e) are blurred or affected by glare, which diminishes their evidentiary value. Figure composition should be improved to ensure clarity and professional presentation.

We have updated the figures as suggested

2. Yield data are provided only for UV resin substrates. No yield or reproducibility analysis has been reported for the more relevant biodegradable substrates (e.g., paper, PHBV), which undermines claims of process robustness.

We are unsure about the scientific merit of this; there is no obvious reason why the yield and reproducibility would be different, as it is essentially the same process onto similar materials. Furthermore, the yield at this point is mostly a function of the cleanliness of the processing environment we have access to (as can be seen where yield drops rapidly at resolutions below 25 μm – where dust contamination becomes a major concern). Improving upon this beyond the high level of reproducibility we have already demonstrated (through the production of large quantities of devices with multiple different types and repeatable performance) would not be of substantial scientific interest to a reader, and in any case would be obviously improved in a dedicated industrial setting. We routinely produce IDE devices on PHBV and paper and, as expected, the yield/reproducibility remains roughly the same – with close to 100 % yield at 100 μm resolution and a rapid fall off in yield below 25 μm . We updated the manuscript just to state this.

In addition, the overall writing quality needs improvement, as the manuscript still contains numerous basic errors that affect readability.

Whilst no specific examples were given by the reviewer, we have read the manuscript carefully for errors.

3. The scalability claim is supported only by a 15 × 30 cm visual demonstration. Quantitative mapping of thickness and sheet resistance across large areas is needed to substantiate uniformity and reproducibility.

In our view, 15x30cm is a reasonably large size for an academic journal and far greater than many examples in the literature (which tend to be around an order of magnitude smaller!). Additionally, there are not many commercial applications of PCBs that have a larger size than this. We are unsure how beneficial it will be to go to larger substrates as this requires production level equipment. There is little scientific merit, this is just further validation of our approach and more suited to a commercialisation strategy.

In terms of uniformity and reproducibility, we have added a line to state that even in our early, proof-of-concept stage, shows less than 5% variance across the sample size and across multiple samples. We used a profilometer and 4-point probe.

4. Evidence for benign degradation remains insufficient. Visual soil tests alone cannot prove safety. Chemical analysis of degradation products (e.g., ICP-MS, SEM-EDX) and ecotoxicity validation (e.g., seed germination) are required.

The author provides an interesting point; however, we strongly feel this is outside the scope of this work. Undertaken a chemical analysis using mass spectrometry or a seed germination study are papers in themselves.

To satisfy the reviewer, we have modified the introduction and referenced literature which discussions the degradation of zinc in soils.

However, we have added the following text in section 5.3, which we feel is a valuable addition and leaves it to readers from an environmental science perspective to consider further studies

“Future studies can focus on the chemical analysis of degradation products using techniques such as ICP-MS, SEM-EDX) and/or ecotoxicity validation using tests such as seed germination”

5. Comparisons to FR-4 PCBs are overstated. Literature tables (Table S4) are not a substitute for direct benchmarking. High-frequency signal integrity tests on the fabricated PCBs remain essential.

This is an interesting point, but outside the scope of this paper as the circuits described in this paper are simple low frequency electronic circuits. HF signal integrity analysis is needed when signal frequencies increase to 100's of MHz, which is far out of the range of this work. HF integrity tests are more for e.g PCIe connections or RF circuits. So, to satisfy the reviewer, we have added text into the SI stating that HF integrity tests will be needed to evaluate the performance for HF circuits.

6. Heater stability was tested only for 60 minutes. No thermal cycling experiments were performed to evaluate long-term reliability under repeated stress, which is a key failure mode.

We think the reviewer is wrong here or perhaps not experienced in this area. Thermal cycling is not a 'failure mode,' it is a stress factor which can be used to create failure modes. It is not a particular interesting test; however, we conducted JESD22-A104 thermal cycle tests and saw limited variation in sheet resistance/performance of the heater and so the heater pass the testing standard. We have added this as text to the SI.

We think 60 minutes is reasonable stress test for early-stage results in a journal paper; and it is unclear what tests the reviewer specifically wants us to do. We have added text stating a thorough reliability test at an accredited test house is needed to evaluate the long-term stability of this before commercialisation of this technology. A detailed reliability study should be undertaken on a final product close to commercial market; this work isn't relevant for an early-stage proof of concept paper.

7. Recycling experiments show functional component recovery, but recovery efficiency is not quantified. Reporting the percentage of successfully retrieved components is critical for evaluating circularity.

Quantifiable data was already in the earlier manuscript. However, we have highlighted the text again for the reviewer to see. 100% of components were recovered and used for a remanufactured circuit with only minor change in performance.

8. Several plots central to degradation claims (e.g., Figure 9c) are presented without error bars. Proper statistical treatment with multiple trials ($n \geq 3$) is necessary to validate the reported trends.

This has been updated

9. Humidity tolerance is reported only qualitatively. Quantitative assessment of circuit performance under varying humidity (e.g., standby current, noise levels) is required to establish operational limits.

We are unsure about the scientific merit of this; this is just further validation experiments which doesn't really provide any useful information for an academic audience. However, this was already measured in earlier work and the following text has now been added

"Standby current didn't change under varying humidity. The noise levels remained lower than the measurable noise from our experimental setup."