Printed technology as a comprehensive paradigm for circular economy of electronics

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The increasing demand for high-performance electronics and sustainability challenges, requires innovative recycling technologies. This study explores the potential of fungal microorganisms in degrading conductive inks, promoting their circular economy integration. Taking advantage of the ability of fungi to process complex compounds, seven fungal strains—*Trametes versicolor*, *Funalia floccosa*, *Pycnoporus cinnabarinus*, *Aspergillus niger* HM81, *Aspergillus versicolor* HM30*, Scopulariopsis brevicaulis* HM03 and *Thielavia* sp. HM70—were tested to degrade copper-based conductive papers (CPs) for 60 days. Post-incubation assessments include gravimetric approach, oxidoreductase enzyme production, attenuated total reflectance-Fourier transform infrared (ATR-FTIR) and X-Ray photoelectron spectroscopy (XPS). The results revealed that all the strains reduced the weight by more than 50% and modified the cellulosic composition, being *P. cinnabarinus* the strain that showed the best results with 77.8% reduced weight. This technique significantly enhances the recyclability of high-performance, yet polluting materials, reducing their environmental impact, and minimising electronic waste.

With the rapid advancement of technology, our society faces new and worrying challenges related to carbon footprint throughout the product life cycles. Such challenges are particularly critical in electronics, a field that has expanded significantly over the last century and relies on high-energy processes and non-sustainable materials, such as FR4 substrates, toxic etching chemicals, and lead-based solders[1]. Furthermore, most of the population is not aware of the possibilities of recyclability and reusability of the electronics components leading to 57.4 Mt of e-waste in 2021 to grow to 74.7 Mt by 2030 with only 17.4% of Waste from Electrical and Electronic Equipment (WEEE) properly treated and recycled in 2019[2].

To face this situation, researchers have been investigating novel ways of producing electronics with a much lower carbon footprint than silicon (Si)-based technology. In this sense, printed electronics have arisen as a promising alternative thanks to the use of low energy processes, the fact that electronic components can be produced on thin-film substrates and the possibility of employing responsible materials. This trend towards printed electronics technologies is justified for several reasons since it eliminates the need of using expensive lithographic processes, high vacuum processing and plasma etching, commonly used in Si-based technology. These traditional methods generate a negative environmental impact, so by eliminating them, printed electronics significantly reduces waste production during the manufacturing process[3].

Various techniques, such as microcontact printing, nano printing, screen printing, and drop-on-demand, enable the development of conductive devices using inks that tend to be more sustainable, such as organic polymer-based and carbon-based inks. Moreover, these inks can be printed on eco-friendly substrates, like paper and glass, resulting in cost-effective products[4–6]. However, these more sustainable materials often limit the performance and durability of the devices.

To balance sustainability and functionality, researchers have focused on combining metallic nanoparticles (typically Ag) with other responsible materials. Recently, conductive materials have been developed using non-polluting substrates along with gold, silver, and copper nanoparticles (NPs) as conductors. These metallic inks offer greater conductivity than inks made from polymer bases[7], thereby enhancing the performance of printed electronics. This development has led to the creation of flexible electronic circuits printed on paper, known as conductive papers (CP). Paper-based printing technology with conductive nanoparticle inks offers advantages like substrate accessibility, low cost, flexibility, and biodegradability[8], contributing to the production of "green economy" products—devices that are both highly efficient and biodegradable, ultimately adding value and sustainability.

The main recycling technique for printed-electronics is solvent-based extraction. Nevertheless, recycling conductive inks, especially those based

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on metallic nanoparticles on paper substrates, poses significant challenges. The sintering process, which is required to enhance the conductivity of metallic inks, creates an irreversible fusion of nanoparticles, making it difficult to recover the metal while retaining its original conductive properties[9]. Current recovery methods, which mainly involve dissolving the ink, have been proposed for silver-based circuits[10–13]. However, similar strategies for copper-based inks are less explored. Indeed, it has been proved that the recycling process that was effective for silver ink did not work for copper ink[14]. This is primarily because copper is more difficult to recover and reuse due to its susceptibility to oxidation and the complex processes needed to obtain it in a pure, conductive state after use. It is important to note that in these studies, the substrates are not paper based, highlighting the added difficulty of applying these methods to more sustainable substrates like paper. Given that the recycling of metallic inks, particularly those based on copper and used on paper substrates, remains relatively unexplored and has not been effectively implemented to our knowledge, it is essential to develop new end-of-life (EoL) strategies for these materials. Addressing these recycling challenges is crucial for managing waste and supporting the principles of a circular economy throughout the entire life cycle of electronic products.

The most common strategies for disposing of printed devices when recycling is not feasible, are incineration and landfill. However, alternatives like mycoremediation could play an eco-friendly role and improve the circular economy compared to incineration or landfill in life-cycle-analysis, by sustainably removing part of the generated waste. Mycoremediation helps to mitigate greenhouse gas emissions and decrease the negative impact and space required for landfills. Additionally, mycoremediation strategies could present advantages in terms of energy consumption, as the fungus can use the residue as a source of carbon and energy for their growth. Several research has demonstrated the powerful biodegradation capabilities of fungi, which can effectively break down complex polymers such as lignin, cellulose, and other recalcitrant compounds like polycyclic aromatic hydrocarbons, synthetic dyes and even plastics due to their potent mechanisms to produces enzymes[15]. These findings highlight the potential of fungi in environmental remediation and industrial biotechnology since they can be employed to degrade environmental pollutants and process

waste. In particular, the fungi from the groups of white rot and ascomycetes have shown remarkable efficiency in decomposing these tough substances[16–22]. Exploring how fungi break down complex chemical structures, this study introduces a method to provide a circular economy-compatible EoL solution for metal NPs that are traditionally difficult to recycle. Biodegradation process provides a promising alternative to conventional recycling methods, which are often energy-intensive and result in a degradation of the functional properties of recycled materials. Additionally, the cost is relatively low because, although some facilities are required to cultivate the fungus, the operational needs for its growth are neglectable. Furthermore, scaling-up the system could be done in a solid bioreactor, which would involve an initial cost but not a significant operational cost[23,24].

In this work, we propose an innovative EoL process for the utilization of copper NPs on paper substrates, within a circular economy context. Hence, the aim of this initiative is the implementation of mycoremediation strategies analysed by spectroscopy techniques in order to reduce the environmental impact of post-used printed electronic devices. The process involves transforming the copper CPs with fungal microorganisms, thereby reducing their toxicity, and converting them into a less harmful material that can be reintegrated into the circular economy. To evaluate the effectiveness of the biotechnological solution techniques such as gravimetric analysis, production of oxidoreductase enzymes, attenuated total reflectance-Fourier transform infrared spectroscopy (ATR-FTIR), X-Ray photoelectron spectroscopy (XPS) and scanning electron microscopy (SEM) will be applied.

Results
Weight reduction of CP
Figure 1 shows the percentage of weight reduction of the CP after 60 days of incubation. The results show that the weight of the CP decreases in three months, more than half of its weight, for each of the strains except for *A. versicolor* that is slightly lower of 50%. However, *T. versicolor* and *P. cinnabarinus* reached a weight reduction of 75.4% and 77.8%, respectively.

Oxidative enzymes production in presence of CP
The test of enzymatic activity production in the presence of the CP is presented in Table 1. The obtained data show that the selected enzymatic
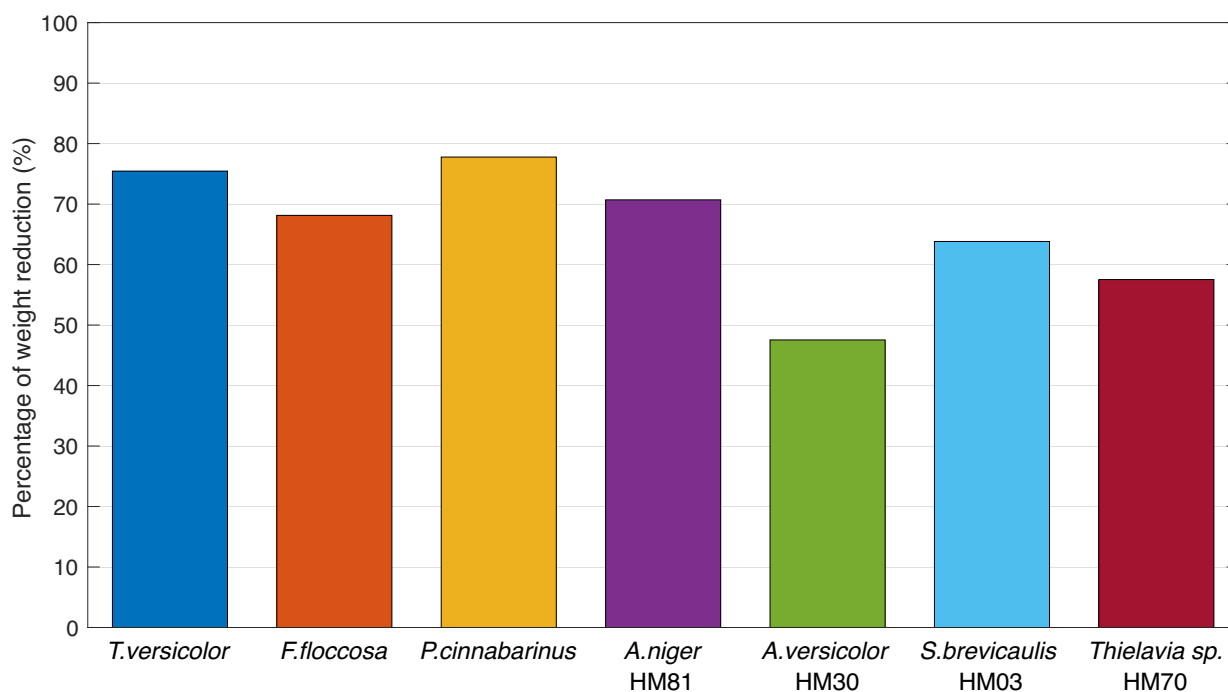

Fig. 1 | Percentage of weight reduction (%) of the CP after 60 days of incubation with the selected fungi. Error bars indicate the standard error of the three replicates ($n = 3$). The percentage of weight reduction was lower than 0.05% and in consequence not represented.

Table 1 | Heat map of the extracellular oxidative enzymes produced by the selected fungi (green: positive result, red negative result)

	Laccase		Peroxidase		Tirosinase		Polyphenol oxidase	
	24 h	48 h	24 h	48 h	24 h	48 h	24 h	48 h
Trametes versicolor	red	green	green	green	red	red	green	green
Funalia floccosa	red	green	red	red	red	green	red	red
Pycnoporus cinnabarinus	red	red	red	green	red	red	green	green
Aspergillus niger HM81	red	red	red	red	red	red	red	red
Aspergillus versicolor HM30	red	red	red	red	red	red	red	red
Scopulariopsis brevicaulis HM03	red	red	red	red	red	red	red	red
Thielavia sp. HM70	red	green	red	red	red	red	red	red

activity was only detected in 4 of the 7 fungi used, *T. versicolor*, *F. floccosa*, *P. cinnabarinus* and *Thielavia sp.* HM70. Thus, *T. versicolor* showed laccase, peroxidase and polyphenol oxidase activity; *F. floccosa* showed laccase and tyrosinase activity. Peroxidase and polyphenol oxidase activity were detected in *P. cinnabarinus*, while *Thielavia sp.* only showed laccase activity.

Changes in the composition of CP by ATR-FTIR

Figure 2 shows the different spectra obtained using ATR-FTIR. The peaks obtained provide important information regarding the chemical composition of the sample. Thus, the spectrum obtained is representative of a characteristic spectrum of cellulose, being able to locate the vibration bands of the C–H$_2$ bonds corresponding to the peaks at 2914 cm^{-1} (asymmetric vibrations) and 2847 cm^{-1} (symmetric vibrations). There is also a peak at 1259 cm^{-1} that would correspond to the C–H bond and a peak at 1160 cm^{-1} that corresponds to the C–O–C bond[25]. In addition, a peak appears at 1730 cm^{-1} that is related to the ketone C = O bonds present in hemicellulose. Finally, the absorption bands at 718 and 1463 cm^{-1} correspond to the reaction between the silane functional group (SiH$_4$) and polyethylene[26]. Data shows that the peaks corresponding to the cellulose were maintained, as well as the vibration bands of the C–H$_2$ bonds (2914 cm^{-1} and 2848 cm^{-1}). However, bands at 1160 cm^{-1} and 1259 cm^{-1}, belonging to the C–O–C and C–H bonds, disappeared after fungal treatment with *T. versicolor*, *F. floccosa* and *P. cinnabarinus*, and a new band corresponding to the C–O bond appears (1032 cm^{-1}). Furthermore, for these three basidiomycetes, a broad band was observed at 3300 cm^{-1}, which regularly appears when measuring solid samples containing water. Now, it can be verified if this band belongs to water by checking if it also has a band between 1630 cm^{-1} and 1650 cm^{-1}, as occurs in these three samples[27]. Finally, peaks representing the reaction between silane and PE (718 cm^{-1} and 1463 cm^{-1}) are still present in all samples, while the band corresponding to C = O bonds (1730 cm^{-1}) disappears.

Oxidative changes in the copper NPs of the CP

The spectra obtained by XPS spectroscopy (Fig. 3) for the analysis of copper indicates the presence of this metal in the conductive paper after 60 days of incubation for the different fungal strains. In the control, this metal was found in the form of CuO since the presence of the four characteristic peaks was observed (933.6 eV; 942.6 eV; 953.4 eV and 961. eV). After treatment with *T. versicolor* a decrease in the intensity and binding energy of the 4 peaks was observed when it was compared with the control. However, the treatment with *A. niger* and *S. brevicaulis* showed the appearance of only two peaks, (931.1 and 951.4 eV for *A. niger* and 931.6 and 951.4 eV for *S. brevicaulis*) at lower intensity, which could correspond to the presence of this metal in the form of cuprous oxide (Cu$_2$O) (Fig. 3a, d)[28,29]. Finally, no trace of this element was observed for the rest of the samples analysed.

Scanning electron microscopy (SEM)

The capacity of the fungi to cover the CP during the incubation time is illustrated in Fig. 4. The pictures analysed by SEM showed the hyphal growth on the surface of the CP by *T. versicolor* (Fig. 4a) and *F. floccosa* (Fig. 4b). These two fungi exhibited a tubular hyphae filamentous network fully covering the surface of the CP. The mycelia growth on the surface might have facilitated the access to the CP and caused the appearance of the broken and damaged areas that are shown in the micrographs. Furthermore, in Fig. 4b, the layer of mycelia showed a thicker layer on the profile of the CP.

Discussion

The obtained results show that *T. versicolor* and *P. cinnabarinus*, basidiomycete fungi belonging to the white rot group, could reduce more than 75% of the weight of these conductive papers. This eco-physiological group of white rot fungi is one of the few microorganisms capable of completely degrading polymers of phenolic origin, such as lignin[22]. For this reason, the use and application of white rot fungi for the purposes of bioremediation of contaminants have received considerable attention during the last decades, with *T. versicolor*, thanks to its enzymatic spectrum, being an excellent model for bioremediation purposes[17]. *T. versicolor* is one of the most efficient basidiomycetes in wood degradation, causing the simultaneous decomposition of lignin, cellulose, and hemicellulose thanks to the release of extracellular enzymes such as laccases, peroxidases and polyphenol oxidases, in combination with the intracellular detoxification system of P450 enzymes, that are capable of degrading these polymers[16,20]. Like *T. versicolor*, *F. floccosa* and *P. cinnabarinus* have great potential in the degradation of this type of compounds, since they are also equipped with oxidative enzymatic systems including laccases and tyrosinases in *F. floccosa* and peroxidases and polyphenol oxidases in *P. cinnabarinus*, detected in this study.

The rest of the tested fungi in this study, belonging to the group of ascomycetes (*Thielavia sp.*, *A. niger* HM81, *A. versicolor* HM30 and *S. brevicaulis* HM03), have been shown to considerably reduce the weight of conductive papers, being *A. versicolor* HM30 the one that has reduced the lowest percentage (47.54%). However, except for *Thielavia sp.* which shows the presence of only laccases, none have presented the studied enzymes, which can be attributed to the experimental conditions. The studies performed in[30] showed that *Thielavia sp.* has the ability to secrete laccases in liquid media and at temperatures of 35 °C, as is an extremotolerant fungus, and the activity of DyP type peroxidases was also detected for the first time in the study. *A. niger* has been widely used in biotechnology industries, as a commercial cocktail to produce hemicellulases and cellulases, and shows excellent performance for the production of enzymes such as glucanases, xylanases, mannases and pectinases, potential enzymes for the degradation of cellulosic compounds[31].
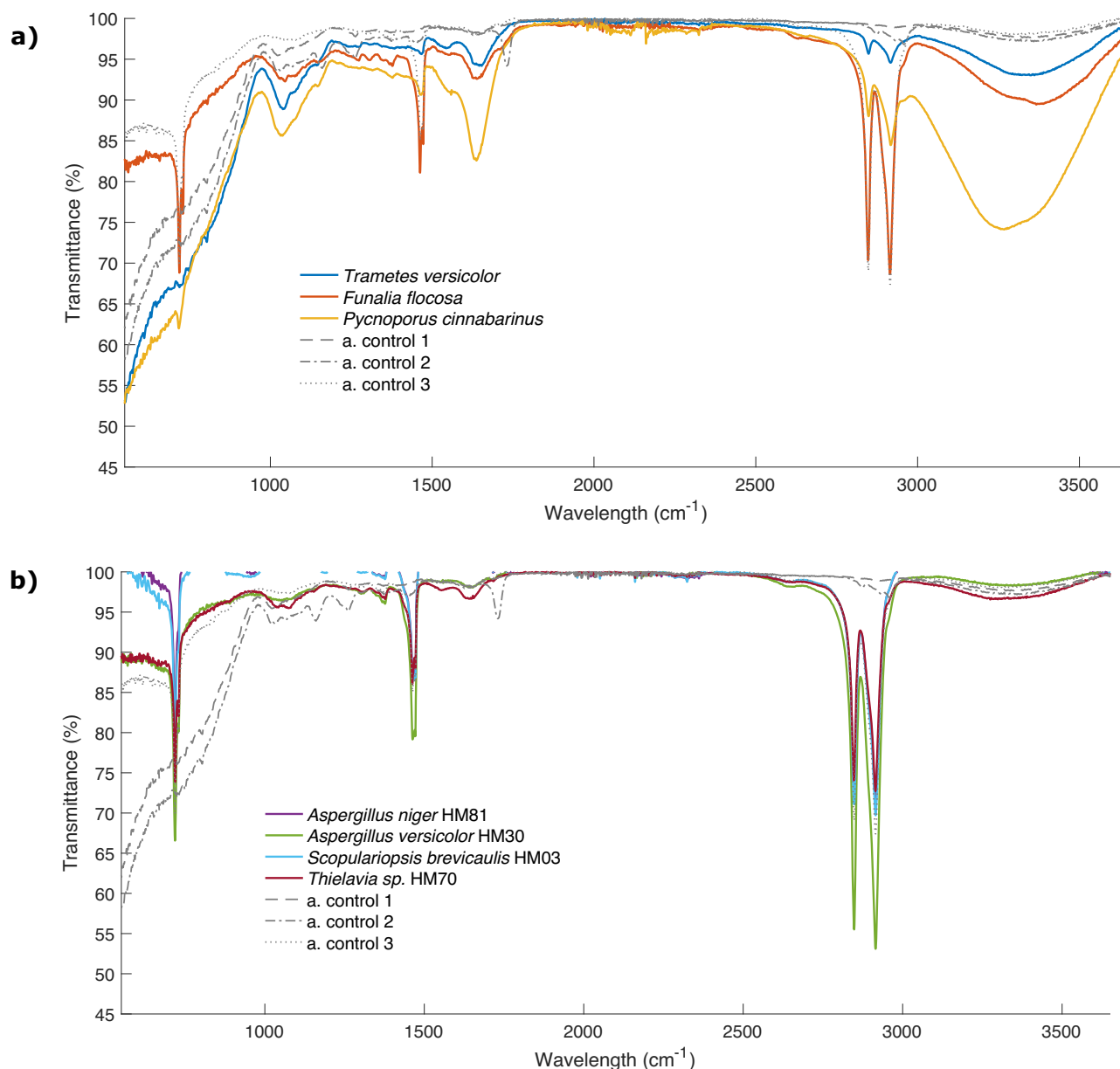
Fig. 2 | ATR-FTIR spectra of CP after 60 days of incubation. a Fungal strains belonging to the group of white rot fungi: *Trametes versicolor* (orange line), *Funalia floccosa* (light blue line), *Pycnoporus cinnabarinus* (yellow line). **b** Fungal strains belonging to the group of ascomycetes: *Aspergillus niger* HM81 (green line), *Aspergillus versicolor* HM03 (purple line), *Scopulariosis brevicaulis* HM03 (dark salmon line), *Thielavia* sp. HM70 (pink line) abiotic control (brown, grey and black line).

Thus, in this study, it has been possible to verify how *A. niger* HM81 has been able to reduce the weight of the papers by more than 70%. Likewise, according to Huang et al.[32], fungi belonging to the *A. versicolor* group, have been shown to be good producers of beta-glucosidases, enzymes involved in the complete degradation of cellulose to glucose, due to their potential to use the most economical substrate as a source of energy and carbon. Relating to the hyaline fungus *S. brevicaulis*, despite not determining the presence of any oxidoreductase enzyme, it managed to reduce the paper weight up to 63%, other studies have identified genes that express endoglucanases along with another series of lignocellulolytic enzymes such as pectinases, hemicellulases, cellulases, peroxidases, polyphenol oxidases; proteolytic and hydrolytic enzymes[26,33].

Regarding the results obtained by ATR-FTIR, it must be taken into account that the reflectance spectra obtained with this technique present some drawbacks for their interpretation, given that the light sent towards the sample is reflected, therefore, depending on the physical state of the

material, the results could show shifted, noisy or even negative peaks[34]. In addition, cellulose spectra, due to its supramolecular structure, present an overlap in the bands, which further complicates its characterization[25]. The existence of a series of changes in the typical bands of a cellulose spectrum can be seen after the alteration of the papers with fungi. Discarding the bands coming from the noise caused by the air (peaks around 3300 cm^{-1} and 1630 cm^{-1}), for all the fungal samples, the bands corresponding to the CO–C (1160 cm^{-1}), C–H bonds disappeared (1259 cm^{-1}) and C = O (1730 cm^{-1}), the latter corresponding to the non-conjugated ketone bonds present in hemicellulose, giving rise to the interpretation that the loss of these bonds is attributed to modifications in the chemical structures in the composition of conductive papers, caused by fungal enzymes. In addition to this, in the spectra of the basidiomycetes strains a new band was observed corresponding to C–O bonds (1032 cm^{-1}) that do not appear in the rest of the fungi and may be a product of the activity of oxidoreductase enzymes identified in this study. Lastly, the absorption bands at 718 and 1463 cm^{-1}
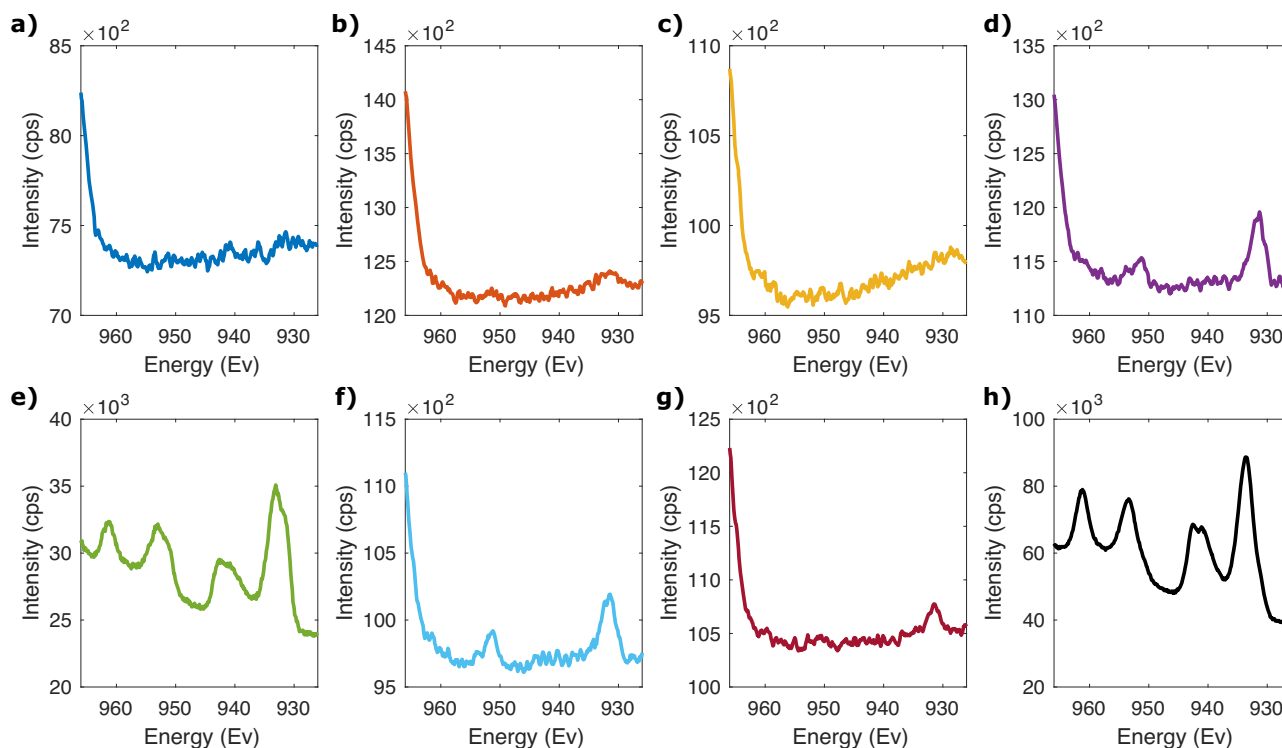
Fig. 3 | XPS spectra of the CP after fungal treatment for copper (60 days). a *Trametes versicolor,* **b** *Funalia floccosa,* **c** *Pycnoporus cinnabarinus,* **d** *Aspergillus niger* HM81, **e** *Aspergillus versicolor* HM30, **f** *Scopulariopsis brevicaulis* HM03, **g** *Thielavia sp.* HM70 and **h** abiotic control.
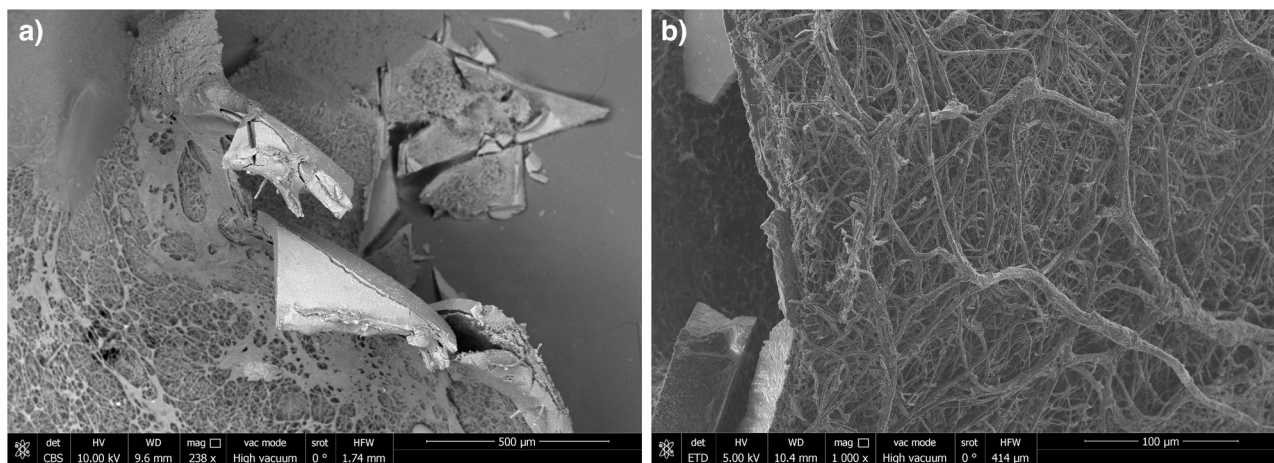

Fig. 4 | SEM images. a General view of the hyphae of *Trametes versicolor* and **b** *Funnalia floccosa* on the surface of the conductive paper after 60 days of incubation.

corresponded to the reaction between the silane functional group (SiH_4), widely used as a coupling and adhesion agent, and PE, also used to form adhesive bonds, remain in the spectra of all fungi, showing that progress must be made in this direction to find mechanisms that facilitate the complete degradation of materials that require this type of elements in their manufacture.

The XPS experiment revealed a decrease of the agent cuprous oxide (Cu_2O). This metal reduction could be mediated by some oxidoreductase enzyme caused by the fungal activity or by a change in the pH of the medium, because of the production of organic acids. For *A. versicolor*, no change was detected in the copper oxidation state of the samples, while for the rest of the samples, the presence of Cu was not directly detected in the analysed area, which can be due to the activity of mobilisation mechanisms to the cell wall such as biosorption or bioaccumulation cannot be affirmed. It

has been reported that the abilities of some fungi, including those of the *Aspergillus* genus, can bioaccumulate copper. That means that these microorganisms are not able to degrade copper but can perform a biosorption process and, in some cases, incorporate it into their biomass, contributing to the removal of the metal from the environment[27,35]. The bioremediation process also includes biotransformation of the target compound, which entails the modification of the chemical structure by enzymatic or non-enzymatic mechanisms. A change in the valence of a metal involves either the reduction or oxidation of the metal by the fungus. This change in valence is a key aspect of biotransformation because it modifies the chemical structure and properties of the metal, often making it less harmful and more stable in the environment. However, with the available data it is not possible to correlate the state of Cu oxidation to the fungal activity, and if the fungal action is primarily affecting the surface of

the substrate used for the printed electronic devices. Furthermore, the particles of a porous or solid surface that have the capacity to absorb and retain liquids on a surface can have important influences on the position, energy and shape of the photoelectron. To elucidate the sorption process by fungi other techniques like Transmission Electron Microscopy Coupled with Energy-Dispersive X-ray Spectroscopy (TEM EDX) should be implemented, as a consequence of changes in the electronic structure of small aggregates[36]. Another factor to point out is the imprecision of the exposed surface using this technique or the possibility that this part of the mycelium was moved while the hyphae were growing on the surface. Even with these questions remaining, it is highly probable that fungal bioaccumulation can not only contribute to reducing copper contamination but also generate a copper-enriched biomass that presents promising opportunities as a reusable composite material in future printed electronic devices.

SEM proved that *T. versicolor* and *F. floccosa* effectively colonised and degraded conductive paper (CP), with visible structural damage indicating active degradation. Differences in hyphal thickness between the fungi suggested varying degradation strategies, which can potentially affect the degradation rate. Therefore, the roughness of the hypha might be associated to biosorption process. It has been previously reported for *P. oxalicum* strain Z6-5-1, where SEM images showed a thicker hypha in the presence of manganese residue[37]. In the case of Cu sorption, melanin produced by fungi has been reported as an efficient biosorbent[38]. *T. versicolor* and *F. floccosa* showed a laccase production and this activity has been correlated with production of melanin[39]. Hence, these could be involved in mechanisms of copper biosorption since it was also observed in XPS results for *T. versicolor*.

Ultimately, understanding the challenges of fungal degradation mechanisms is crucial not only for optimizing the process at a micro-level, but also for evaluating its broader implications when considering its adoption as a sustainable waste management strategy, particularly when compared to existing methods. Compared to traditional methods like incineration and landfill disposal, fungal biodegradation demands far less energy, with its primary energy requirement being establishing a suitable environment for their enzymatic decomposition of the materials. In terms of cost, fungal biodegradation can also offer significant savings. Furthermore, while a comprehensive cost analysis is needed, it is likely that fungal biodegradation could offer a more economically viable solution than incineration's high operational costs and the long-term expenses associated with landfill maintenance. While the initial setup costs for fungal biodegradation facilities may be high, the operational costs are generally lower due to the self-sustaining nature of fungal processes. Since fungi can grow and reproduce on their own, reducing the need for costly inputs such as chemicals or machinery. Additionally, the byproducts of fungal biodegradation can often be used as valuable resources, which can even compensate for the costs associated. Moreover, unlike solvent dissolution, which efficiently recovers certain metals but uses hazardous substances and generates chemical waste, our approach eliminates the need for such substances. This minimises risks to human health and the environment. However, our fungal biodegradation method is slower than chemical dissolution and incineration and may require extended incubation times and further optimization to achieve complete degradation. Scalability is another important factor to consider when comparing fungal biodegradation with existing methods. Fungal biodegradation can be easily scaled up or down depending on the volume of waste materials to be processed. This flexibility allows for efficient waste management in a variety of settings, from small-scale operations to large industrial facilities. In contrast, traditional methods such as incineration or landfilling may be limited in their scalability since it depends on the infrastructure already made. Finally, fungi are natural decomposers that play a crucial role in nutrient cycling and ecosystem health. Making use of this potential of fungi for waste management, we can reduce the amount of waste sent to landfills or incinerators, leading to lower greenhouse gas emissions, and reduced environmental pollution resulting in a lowered environmental impact compared to traditional methods.

In conclusion, this study examines how seven fungi from two major groups, Basidiomycetes and Ascomycetes, could enhance the sustainability of conductive papers used in the electronic through their ability to biodeteriorate them. To this end, this work proposes the use of fungi to find a way to biodegrade these devices and eliminate the heavy metals used in their manufacture, to scale towards more sustainable devices and achieve the "green economy" during manufacturing and after use. The results obtained in this work show that the use of fungi represents a good way to degrade conductive papers, since: i) a decrease in the weight of the papers by more than 50% has been detected in the 7 fungal strains in 60 days, ii) alteration of the composition of the cellulose of these papers was detected, and iii) a change in the oxidation state of the copper present in the samples of *A. niger* and *S. brevicaulis* was achieved. However, although the data obtained in this study are promising, more studies should continue in this way, increasing the incubation time, analysing the hydrolytic enzymes, using complementary techniques to support the obtained results, expanding information such as intracellular enzymes of the cytochrome P450 type, studying the mobilisation of the heavy metals printed on papers and the degradation of other types of compounds identified in this study such as xylan and polyethylene, used with adhesion and coupling functions. Although copper has not been completely eliminated, this method shows great promise. By implementing successive stages of fungal treatment, process efficiency could be improved, ultimately leading to the complete degradation of all the material involved. This approach represents a novel EoL solution for electronics and offers a significant advancement in the field.

Methods

Conductive papers fabrication

Copper oxide ink ICI002HV from Novacentrix (Texas, US), containing 16% fully oxidized copper nanoparticles, was deposited onto Epson Matte photographic paper substrates using inkjet printing. The samples were allowed to dry for approximately 30 min and then sintered using a Sinteron 2010 (Xenon, US) with a single 2 ms pulse at 3.0 kV[40] (Supplementary Fig. 1).

Fungal strains

Seven fungal strains were selected as potential candidates for the degradation of the CP based on their capacities to produce extracellular enzymes involved in lignocellulose degradation: *Trametes versicolor*, *Funalia floccosa* and *Pycnoporus cinnabarinus* belonging to the group of white rot fungi and *Aspergillus niger* HM81, *Aspergillus versicolor* HM30, *Scopulariopsis brevicaulis* HM03 and *Thielavia sp.* HM70 from the group of ascomycetes.

Trametes versicolor, *Funalia floccosa* and *Pycnoporus cinnabarinus*, were obtained from the collection of Dr. Inmaculada García Romera, from the Estación Experimental del Zaidín (CSIC, Granada, Spain) and *Aspergillus niger* HM81, *Aspergillus versicolor* HM30, *Scopulariopsis brevicaulis*, HM03 and *Thielavia sp.* HM70 were obtained from the mycobiota of sewage sludge composting piles[41] and conserved at the culture collection of the Institute of Water Research. The strains were maintained in Petri dishes with Malt Extract Agar (MEA) medium (VWR International, USA).

Fungal pre-inocula

Pre-inocula were obtained using fresh plate medium incubated at 28 °C for 3–5 days. Once the fungi colonised the entire plate, the medium containing the fungus was blended with 80 mL of sterilised distilled water using an Ultra Turrax (IKA, Germany) under sterile conditions. The obtained mixture was used as a pre-inoculum. Each Petri dish was inoculated with 1 mL of the appropriate fungus with the help of a sterile Digralwsky spatula on the surface of the petri dishes containing MEA with 50 µg·mL^{-1} of tetracycline and 50 µg·mL^{-1} of streptomycin, to avoid bacterial contamination[42].

Mycodegradation of CP

The conducting papers were divided and cut into four parts. Each of the sections was sterilised by immersing them in 70% ethanol for 5 min. They were air dried inside a laminar flow chamber to evaporate the ethanol and were preserved inside sterile microcentrifuge tubes. For the mycodegradation experiments, each of the four pieces of the CP were placed on the MEA

previously inoculated with the target fungi (Supplementary Fig. 2). The plates were incubated in an incubator at 28 °C for 60 days, respectively. The experiment was conducted in triplicate. After incubation and prior to the different analyses, the samples were washed with distilled water in order to remove the mycelium of the fungus that had grown on the papers and then they were allowed to dry on absorbent paper to remove excess moisture.

Determination of the percentage weight decrease of CP
To determine variation in the weight of the CP, each of the four sections were weighed before and after the incubation with the selected fungi using a precision scale. Measurements were conducted in triplicates for each piece of CP.

Analysis of extracellular enzyme production
After 60 days of fungal incubation, one quarter of the Petri dish containing one of the four incubated CP was transferred to a new sterile plate to evaluate the extracellular oxidative enzymes produced by the fungi. The presence/absence of laccase, peroxidase, tyrosinase and polyphenol oxidase was evaluated according to the methodology[43]. Changes of colour after 24 and 48 h in the area where the drops were deposited, was considered as the presence/absence of the tested enzyme.

Fourier transform infrared spectroscopy (ATR-FTIR)
The attenuated total reflectance-Fourier transform infrared spectroscopy (ATR-FTIR) technique was used to characterise the CP, as well as to detect possible changes in their composition after 60 days of incubation with the selected fungi. The ATR-FTIR measurements were carried out at the Scientific Instrumentation Centre of the University of Granada, using a Perkin Elmer Spectrum Two spectrometer, equipped with an attenuated total reflectance (ATR) crystal, which consists of a diamond crystal at a fixed angle of 45°. Diamond absorbs infrared radiation from 2300 to 1800 cm^{-1}, so it is not useful in this region; cellulose does not present absorption bands in this range, so using this type of crystal does not represent any limitation[25]. In this way, the scan was carried out with a spectral resolution of 4 cm^{-1} and a wavenumber range of 4000 to 400 cm^{-1}, and the background noise produced by the air was eliminated every 2–3 samples.

X-ray photoelectron spectroscopy (XPS)
X-ray Photoelectron Spectroscopy (XPS) was used to detect oxidative changes in the copper NPs located on the cellulosic substrate after 60 days of incubation with the fungal strains. Therefore, after 60 days of incubation, a quarter of the samples of the CP was washed with distilled water, deposited on a new Petri dish and dried in an oven at 26 °C for 24 h. After that, the section of the conductive paper corresponding to the area in which the copper NPs had been printed was cut out, to study the presence of this element in the samples. The analyses were carried out at the Scientific Instrumentation Centre of the University of Granada using a Kratos AXIS Ultra-DLD Supra Photoelectron Spectrometer. The software XPSpeak 4.1® was used to process the peaks of the XPS spectra.

Scanning electron microscopy (SEM)
Samples after 60 days of incubation were taken from the Petri dishes. The samples were immediately fixed in a 2.5% glutaraldehyde solution[44,45] in 0.05 M cacodylate buffer pH=7.4 for 24 h at 4 °C. Later, samples were three times washed in 0.1 M cacodylate buffer for 15 min at 4 °C. Post fixation was done with 1% osmium tetroxide in aqueous solution for one hour in the dark and at room temperature. After that, the samples were washed in distilled water for 10 min, this wash was repeated three times. Dehydration was performed in an increasing gradient of ethanol (50%, 70%, 90% and 100%) for 10 min and repeated for three times at room temperature. Critical point drying was carried out in an EM CPD300 Critical Point Dryer CO_2 desiccator (Leica Microsystems, Germany) and subsequently, carbon metallization in an Turbo-pumped thermal evaporator EMITECH K975X (Quorem Technologies, United Kingdom). These samples were prepared and examined by scanning electron microscopy to analyse the fungal growth on the CP surface

in the Centre for Scientific Instrumentation of the University of Granada (Spain), using a SEM in a FEI Quanta 400 device (30 kV, 3.5 nm resolution).

Data availability
All data supporting the findings of this study are available within the article and its Supplementary Information files. Raw data is available from the corresponding authors upon request.

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T.R.M: methodology, writing-reviewing, and edition; S.G.G.: conceptualization, methodology; A.B.: methodology, investigation, formal analysis, writing original draft preparation; V.T.: methodology, investigation, writing-reviewing; F.J.R.: formal analysis, writing-reviewing, and edition; E.A.: Conceptualization, funding acquisition, supervision, writing-reviewing and editing; A.R.: Conceptualization, funding acquisition, supervision, writing-reviewing and editing. All authors approved the submitted version.

Competing interests

The authors declare no competing interests.

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