Green Chemistry

PERSPECTIVE

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Cite this: *Green Chem.*, 2026, **28**, 747

Sustainable advancements in fused filament fabrication/fused deposition modelling additive manufacturing for electroanalysis

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This review explores the advancements made toward sustainable practices in the field of additive manufacturing for electroanalysis. The adoption of Fused Filament Fabrication within the field of electroanalysis has allowed the development of unique sensing platforms, but reliance on commercially available conductive filament has limited the field. Through the development of bespoke filament researchers have progressed both the performance and sustainability of the produced filaments, moving towards using recycled polymers and bio-based additives. Key advancements have been made utilising base polymers with improved chemical and electrochemical stability, facilitating the transition away from single-use electrodes. Despite these advancements, critical challenges remain, especially considering the end-of-life processing of these items and the implementation of closed-loop recycling systems. Continued efforts are essential to realise a true circular economy electroanalytical device fabrication.

Received 10th September 2025,
Accepted 27th November 2025

DOI: 10.1039/d5gc04746c

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1. We discuss the strategies used to improve sustainability throughout additive manufacturing electroanalysis, from optimised design and post-processing techniques to the latest developments in bespoke filament production.
2. Additive manufacturing is a rapidly expanding field, and electrochemistry is fundamental behind various applications of significant interest, including sensors, batteries, electrolysers, supercapacitors, and electrosynthesis. The work summarised here should offer inspiration to people in these wider fields that sustainable approaches can often lead to improved performance.
3. Uniquely, we highlight many examples where the use of more sustainable approaches improves the electrochemical performance of filament for electroanalysis. These synergetic findings should encourage researchers to explore these avenues further.

1. Additive manufacturing electrochemistry

Additive manufacturing, also known as 3D-printing, refers to a whole group of manufacturing processes in which material is deposited in thin-layered cross-sections to build a 3-dimensional object in a layer-by-layer fashion.[1–5] Within these wide ranging processes, Fused Filament Fabrication (FFF), also commonly referred to as Fused Deposition Modelling or FDM, has seen a rapid increase in popularity within the field of electrochemistry and in-particular within electroanalysis.[6–11] FFF is a material extrusion based process, whereby thermoplastic filament is melted and extruded through a hot end and forced out of a nozzle of set diameter (commonly between 0.2–0.8 mm).[12–15] As the molten thermoplastic is forced out of the nozzle, the print head moves around the print bed in the *x*- and *y*-axis, drawing the layer predefined in the computer model. Once the layer is complete, the print head moves in the *z*-axis to draw the next layer on top, with this process repeating until the model is finished. The surge in popularity of FFF within the field of electrochemistry stems from a variety of factors, including the low cost of entry, design flexibility, rapid prototyping, commercial availability of materials, and low waste generation.[16–20] Excellent quality FFF printers are readily available on the market for only a few hundred GBPs, making them a cost-effective addition to research laboratories, whilst only consuming typically between 50–250 W h^{-1}. These machines allow users to print bespoke designs in the lab, next to their electrochemical set-up, removing significant time and cost barriers when making new parts. It is also important to recognise the reduction in transport costs and emissions that are saved by bringing manufacturing in-house. Some of the intricate geometries that additive manufacturing can allow (*e.g.* lattices and gyroids) have not been fully explored within the field of electrochemistry. Nevertheless, the ready avail-

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ability of commercial conductive filament for FFF has facilitated its use to create novel electrodes,[11,21,22] electrochemical cells, electrochemical equipment and even simple but useful lab accessories, whereby researchers can simply design their items on their computer and print them immediately within their lab.[23] For example, in this way electrodes of various working diameters can be fabricated and tested within minutes, rather than a long and expensive procedure with external companies. Although the use of commercial filament has allowed researchers to utilise FFF to design unique electroanalytical platforms, significant improvements in the electrochemical performance of the filament were required to make additive manufacturing electrochemistry a viable commercial alternative. In addition to improving the electrochemical performance, the creation of bespoke filament has allowed researchers to realise substantial advances in their sustainability.[24,25] We first define the production methods reported in the literature before exploring these improvements.

2. Filament production

For additive manufacturing to produce electrodes that can be competitive with classical electrodes and become a staple of the electroanalyst's arsenal, the development of bespoke conductive filament was required. Now commonplace within the literature, a plethora of filaments have been described that vastly improve the electrochemical performance compared to commercially available alternatives. Not only do these filaments improve the standard electrochemical responses, but they can also incorporate specific functionalities to meet the requirements of individual electroanalytical applications, further optimising the performance. To create these filaments within a research environment, two approaches have been reported: (1) solvent mixing, and (2) thermal mixing, both summarised in Fig. 1.

The solvent mixing methodology follows a general protocol of (1) dissolving the base polymer material within a solvent, (2) adding the desired additives/fillers under stirring, (3) casting the solution and allowing solvent evaporation, (4) drying and granulation of the resultant composite, (5) extrusion into filament. This methodology is widely accessible to laboratories around the world as the production of the conductive polymer composite uses standard laboratory glassware and equipment, and reasonable quality filament extruders can now be purchased commercially at competitive prices. Although accessible, the use of this method can result in poor quality filament due to the low shear mixing techniques used (typically a magnetic stirrer bar). This low shear mixing can cause poor dispersion of fillers (*i.e.* carbon morphologies) throughout the polymer matrix resulting in agglomeration of particles and therefore poor printability and inconsistent conductivities. When considering the environmental impact of this methodology, the production of filament in this way can be a low energy cost process by using a small heating mantle (if required) to dissolve the polymer, and it can be tailored to produce the exact amount of filament desired for the final application to reduce plastic waste. As such, using this method on a small scale can be beneficial. Clearly a major environmental concern with this method is the use of large quantities of solvents, typically hundreds of mL to make tens of grams of filament, to dissolve the polymer and disperse the fillers. The majority of the literature on bespoke filaments has concentrated on using PLA as the base polymer, with various solvents being reported, including xylene,[26,27] dimethyl formamide (DMF),[28] toluene,[29] dichloromethane,[29] and one of the most popular being a mixture of chloroform and acetone in various ratios.[30–36] All of these solvents are hazardous toward human health in some way, ranging from causing dizziness and irritation to severe kidney, liver and lung damage in extreme cases. In terms of the environmental impact, all of these solvents are volatile and contribute to local air pollution, with all having the potential to cause significant damage to aquatic life. On top of these crucial factors, this method of composite preparation can be slow, with many reports stating times of well over 1 hour for simply dissolving the polymer, followed by similar times mixing in the fillers, and normally and overnight evaporation of solvent/drying of the composite. It is important to relate these potential effects back to the scenario of filament production, where in a lab-environment it can take a few hundred millilitres of solvent to create only 25–50 g of composite and production scales can fit adequately into a researcher's routine. Although this is fine for creating enough filament to print electrodes for an academic publication, scale-up of these methods to meet industrial and commercial needs is not advisable.

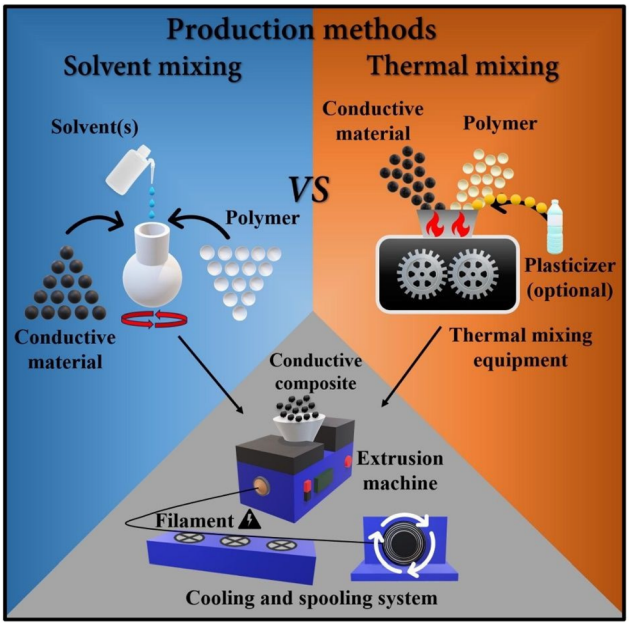

Fig. 1 Schematic representations of solvent mixing methodology and thermal mixing methodology to create bespoke conductive filament for additive manufacturing. Reproduced from ref. 24. Copyright Elsevier 2023, Creative Commons License CC BY 4.0.

Conversely, thermal mixing (also referred to as melt compounding) offers very different characteristics. In this method, all components of the desired filament are added to a heated chamber that is set to the desired temperature for the polymer used and fitted with rotating blades (such as Banbury rotors) to mix the composite under high-shear conditions. It should be noted that other methods of thermal mixing have been reported that use minimal low-shear mixing in an open system,[37,38] however the variation and effect this has on the produced filament is yet to be established. Once added to an automated machine the components are mixed for a set amount of time, with 5 minutes being the most commonly reported in the literature.[39] After mixing, the polymer composite is removed from the machine and allowed to cool, before being granulated and ready for filament extrusion. As such, the total processing time for filament production using this method can be less than an hour, a significant decrease on the time required for the solvent methodology. In general, these machines require a significant initial outlay, along with increased costs in running the devices and appropriate extraction due to the potential release of volatile organic compounds among other risks when melting polymer compounds.[40,41] Clearly, the removal of all solvents from the production process is a significant environmental benefit to thermal mixing, with the potential powering of equipment through green energy sources another noted advantage. Additional potential causes for environmental concern are the necessity for additional polymer use when cleaning equipment as to not contaminate future samples. Ideally, this waste will be recycled to create a closed-loop system, but if not, appropriate correct waste disposal should be a priority.

3. Materials

Within the production of bespoke conductive filament, there are typically three material constituents: the base polymer, the additive such as a plasticiser compound, and the conductive filler. We will now explore these components and highlight the advancements that have been made in these areas for additive manufacturing electroanalysis.

3.1. Base polymers

Due to the production of bespoke filament in-house, in recent years additive manufacturing for use in electroanalysis has seen an increase in the number of base polymers reported. The commercially available conductive filament utilises poly (lactic acid) (PLA), and as such there has been a significant number of reports of bespoke PLA filaments that directly benchmark against this. However, there have also been reports of conductive poly(ethylene terephthalate glycol) (PETg),[42,43] poly(propylene) (PP),[44,45] and thermoplastic polyurethane (TPU).[46,47] All of these base polymers offer their own unique intrinsic properties that can help ensure that sustainability remains at the forefront when designing electroanalytical platforms. Firstly, if we consider PLA, significant number of publications utilising PLA still base a large proportion of their sustainability claims on the biodegradability of the polymer.[48–52] Although technically true to the definition, whereby biodegradable materials undergo degradation to water, carbon dioxide, methane, basic elements, and biomass through the action of living organisms,[53] PLA can only undergo this transformation under industrial composting settings, not within a natural environment. In fact, it has been shown that within a marine environment, PLA showed no significant degradation over 428 days, similar to that observed for oil-based plastics such as PP and PET.[54] Without adequate infrastructure, simply replacing oil-based plastics with PLA[55] is not the answer some reports would lead you to believe. Coupled with the ready ingress of solutions into PLA and its poor chemical stability,[56] all PLA-based electrodes are essentially single-use items. To somewhat offset these issues, the use of recycled PLA as the base polymer has gained significant traction, since the seminal paper from Sigley *et al.*,[57] Fig. 2. In this work, the authors used post-industrial recycled PLA (rPLA) waste from coffee pod manufacturing as their base polymer, creating both non-conductive and conductive filament. The conductive filament contained 61.62 wt% rPLA, along with 29.60 wt% carbon black as the conductive filler and poly(ethylene succinate) (PES) as a plasticiser to ensure good printability. Importantly, the final electroanalytical platform was designed in two separate prints, ensuring the non-conductive cell could be readily recycled again into filament. The authors showed that four cycles were possible for the filament before printing was no longer possible. The platform was used for the electroanalytical detection of caffeine within tea and coffee samples, meaning a full cycle from coffee pod to coffee sensor was realised, with the authors coining the term "circular economy electrochemistry".

Although a step forward, the problems that render PLA-based electrodes effectively single-use remain and as such researchers have begun exploring other base polymers mentioned above. The first of these reported was the use of PETg, another commonly used filament within standard non-con-

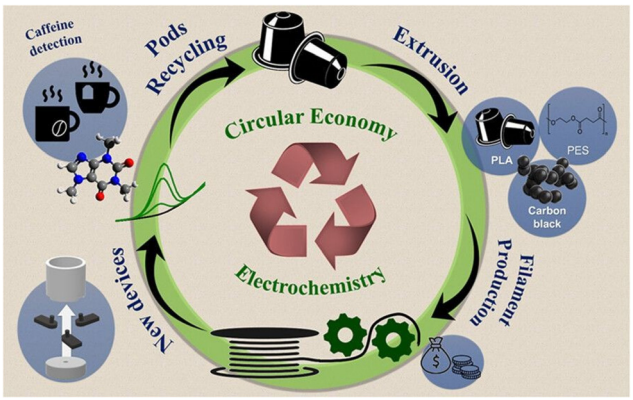

Fig. 2 An overview of the circular economy electrochemistry principles. Image reproduced from ref. 57, copyright American Chemical Society 2023, Creative Commons License CC BY 4.0.

ductive FFF printing. It is known to be relatively easy to print (not as simple as PLA) and less prone to warping than other common printable materials like acrylonitrile butadiene styrene (ABS). It is seen as a compromise between ABS and PLA, bringing the toughness, impact resistance and chemical resistance of ABS, but the ease of printing of PLA. Importantly for the development of electrodes that aim to not be single-use items, PETg is significantly more waterproof than PLA, with it being shown that over a 9 weeks immersion period in water only a 0.3% increase in part weight was observed.[58] In addition to this, PETg offers significant improvements in chemical resistance when compared to PLA, in particular within acidic and basic mediums.[56] First conductive PETg filaments have been reported, utilising recycled PETg prints as the base polymer along with a mixture of carbon materials such as carbon black, multi-walled carbon nanotubes and graphene nanoplatelets as the conductive fillers.[42,43] Within these reports, PETg shows significant improvements in intrinsic properties over PLA. These include showing minimal memory effects or ingress into the polymer matrix,[43] and the ability to be cleaned and re-used for the same application up to 10 times before showing a significant decline in electroanalytical

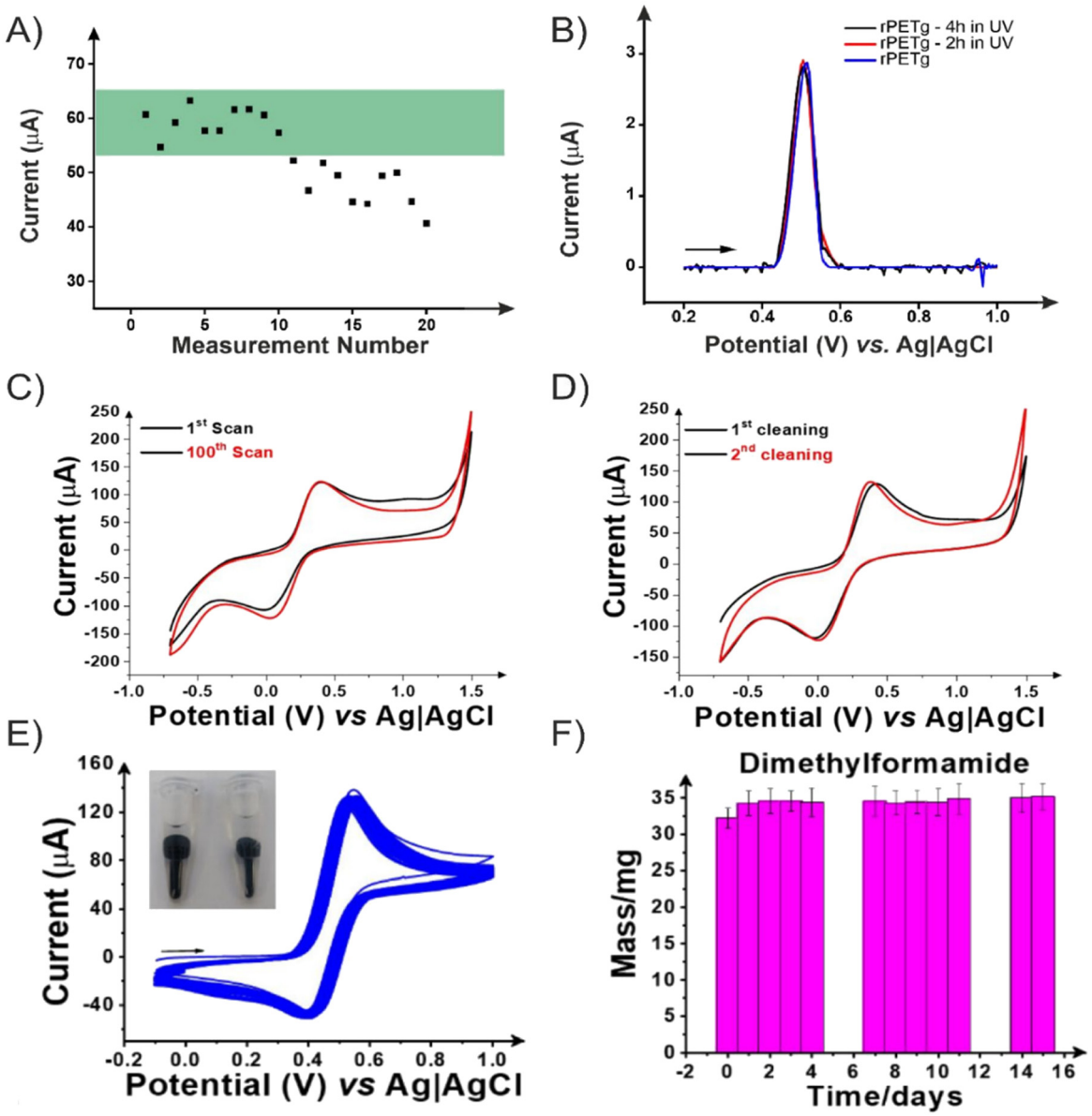

Fig. 3 (A) A plot showing the stability of an rPETg filament over 10 separate measurements and (B) the stability of the DPV response of an rPETg electrode after UV sterilisation. Reproduced with permission from ref. 42, copyright Elsevier 2024. (C) and (D) CV profiles of conductive TPU electrodes in ferricyanide over 100 scans and after cleaning. Reproduced with permission from ref. 47, copyright Elsevier 2024, Creative Commons License CC BY 4.0. (E) CV profiles for 100 scans of ferrocene in DMF using a conductive PP electrode, with an inset image showing the swelling of a PLA electrode (left) *versus* no swelling in a PP electrode (right), and (F) a bar chart showing the stable weight of electrodes for 15 days submersion within DMF. Reproduced from ref. 45, copyright American Chemical Society 2024, Creative Commons License CC BY 4.0.

performance, Fig. 3A.[42] Both of these findings indicate a potential movement away from single-use electrodes which can greatly decrease the amount of polymer waste produced by these electroanalytical platforms. In addition to this, the PETg based electrodes were shown to be sterilisable through alcoholic[43] and UV[42] treatments, Fig. 3B, showing their sterilisability and potential for use within the healthcare, with re-sterilisation and re-use a possible method of reducing waste in this sector.

Also with clear applications within health monitoring has been the development of conductive TPU.[47] TPU is already a filament found within the arsenal of additive manufacturing due to its high flexibility and durability, as well as its resistance to abrasion, oils, greases and solvents, giving it long-lasting performance in harsh environments and conditions.[59,60] Although the majority of precursors for the production of TPU remain petroleum based, there is research on green sources of diisocyanate, polyol, and chain extenders to make the production more sustainable.[60] The sustainable advantages that TPU can present for electroanalysis through additive manufacturing stem from its overall stability when compared to PLA. In work from Oliveira *et al.*[47] they exemplify such stability, showing that TPU based electrodes show no deterioration in the electrochemical signal over 100 scans of cyclic voltammetry, Fig. 3C, demonstrating the effective durability that can be achieved with this base polymer. Additionally, the authors provide evidence that TPU based electrodes can be used, cleaned, and then re-used presenting identical performance, Fig. 3D. This once again shows how TPU-based electrodes have the potential to not be single-use items within this field.

The last change in base polymer explored for electroanalytical applications is the use of PP.[44,45] PP offers high versatility as a base polymer, with significant improvements in chemical resistance, mechanical strength, and durability when compared to PLA.[61,62] Although slightly more challenging to print as PP adhesion to standard commercial print beds is currently limited, PP produces filament with excellent flexibility and requires no additional plasticiser for the addition of up to 40 wt% carbon black filler.[45] Ramos *et al.*[45] presented the first report of conductive PP-based filament for additive manufacturing electrochemistry, showing its application toward electrosynthesis and the electroanalytical determination of colchicine within environmental water samples. The chemical stability of PP is one of its standout intrinsic properties, which the authors showed through 100 scan cycling and immersion testing over 15-days within acetonitrile, dichloromethane, and dimethylformamide, Fig. 3E and F. This stability not only opens the field of additive manufacturing electrochemistry to new areas, such as the green electrosynthesis of new active pharmaceutical ingredients and electroanalysis within organic solvents,[44] but also indicates long-lasting products and a movement away from single-use platforms that plague PLA. PP is also one of the most widely utilised polymers globally, and as such will present significant opportunities for using recycled plastic feedstock.

3.2. Additives

The main additive used to produce conductive filament for additive manufacturing electroanalysis has been a plasticising compound. The addition of these compounds is vital for producing highly filled PLA-based filament that maintain adequate low-temperature flexibility and good quality printing.[63–65] It is important to note that this is an additional advantage of using PP, TPU, and PETg as they do not require this additional plasticiser and therefore do not have the same environmental impact generated by the production and transportation of an additional component. Even so, PLA remains the most popular base polymer used for bespoke filament production, with the first truly flexible versions reported using the addition of poly(ethylene glycol) (PEG)[39,66,67] or poly(ethylene succinate) (PES)[57] as plasticising compounds. A plasticiser is defined by The Council of the International Union of Pure and Applied Chemistry (IUPAC) as "a substance or material incorporated into a material (usually plastic or elastomer) to increase its flexibility, workability, or distensibility".[68] The miscibility of PEG with PLA, and its ability to lower the glass transition temperature of PLA, as well as enhancing the interaction between PLA and fillers meant that these composites gave excellent results.[69–71] Although classed as biocompatible and used widely within pharmaceutical and cosmetic applications,[72] some PEG derivatives have shown cytotoxicity.[73] The production of PEG is through an ethoxylation process, involving acidic or basic catalysts, and can produce some harmful by-products such as ethylene oxide or 1,4-dioxane, which have been shown to be persistent in the environment.[74]

Although only making up a relatively low fraction of the overall composition of filament (5–20 wt%), researchers have reported alternative plasticiser compounds that are fully bio-based. The first report was the use of castor oil at 10 wt% with rPLA and carbon black,[75] which has since been used in multiple reports of rPLA-based conductive filament with carbon loadings up to 35 wt%.[76] Castor oil is a non-edible oil derived from the seeds of the castor plant (*Ricinus communis*), and as such can be collected through mechanical pressing or solvent extraction, with mechanical pressing the preferred option with a sustainability mindset.[77] It is an attractive option due to its widespread cultivation on industrial scales in many countries, low-cost, and its inedible nature meaning its use does not take away from food sources.[78] Since the publication of this seminal paper on bio-based plasticisers within conductive filament production, other bio-based oils have been reported, such as soybean oil[37] and babassu oil.[38] These filaments were prepared in a similar way to the castor oil filaments, utilising 10–15 wt% of oil and thermally mixed to produce the composite for extrusion. All these bio-based oils produce good quality filament, however all aspects of the use of these bio-based oils should be considered when establishing their environmental impact. In terms of global production, soybean oil is produced globally in large quantities with approximately 59 million tonnes produced annually in the last 10 years,[79,80] compared to only around 800 000 tonnes of castor oil annually[81] and

82 000 tonnes of babassu oil.[82] In terms of growth, soybean oil production requires large areas of land and has been linked as a major driver of deforestation, biodiversity loss, and increased greenhouse gas emissions.[83] Babassu oil is only primarily grown in Brazil, where the palms are native and therefore indicates significant transportation emissions if used for filament production globally. On the other hand, castor oil can be produced globally on marginal lands, which reduces competition with food crops and helps prevent deforestation and preserve biodiversity.[84] This highlights how consideration of the whole supply chain is crucial.

3.3. Fillers

Within the production of bespoke conductive filaments carbon allotropes are the most widely used filler for FFF. Of the many options of carbon allotrope, carbon black is the most widely reported. This black powder is most commonly produced through the incomplete combustion of heavy aromatic oils in a furnace,[85] which produces extremely fine particles of around 10–500 nm in size. Alternative carbon allotropes reported include multiwalled carbon nanotubes (MWCNT), graphene, and graphite. MWCNTs are commonly produced through chemical vapour deposition (CVD), which is an energy intensive process that can produce significant CO_2 emissions and release hazardous by-products such as metallic nanoparticles or volatile organic compounds (VOCs).[86] Graphene can also be produced through CVD or through the reduction of graphene oxide, which is cheaper but uses toxic chemicals and can generate a significant amount of waste.[87] Graphite is a naturally occurring conductive form of carbon, however mining operations and the subsequent deforestation, habitat destruction, or soil erosion this can cause should be considered.[88] Alternatively, synthetic graphite can be produced, with the highest purity forms made from petroleum needle

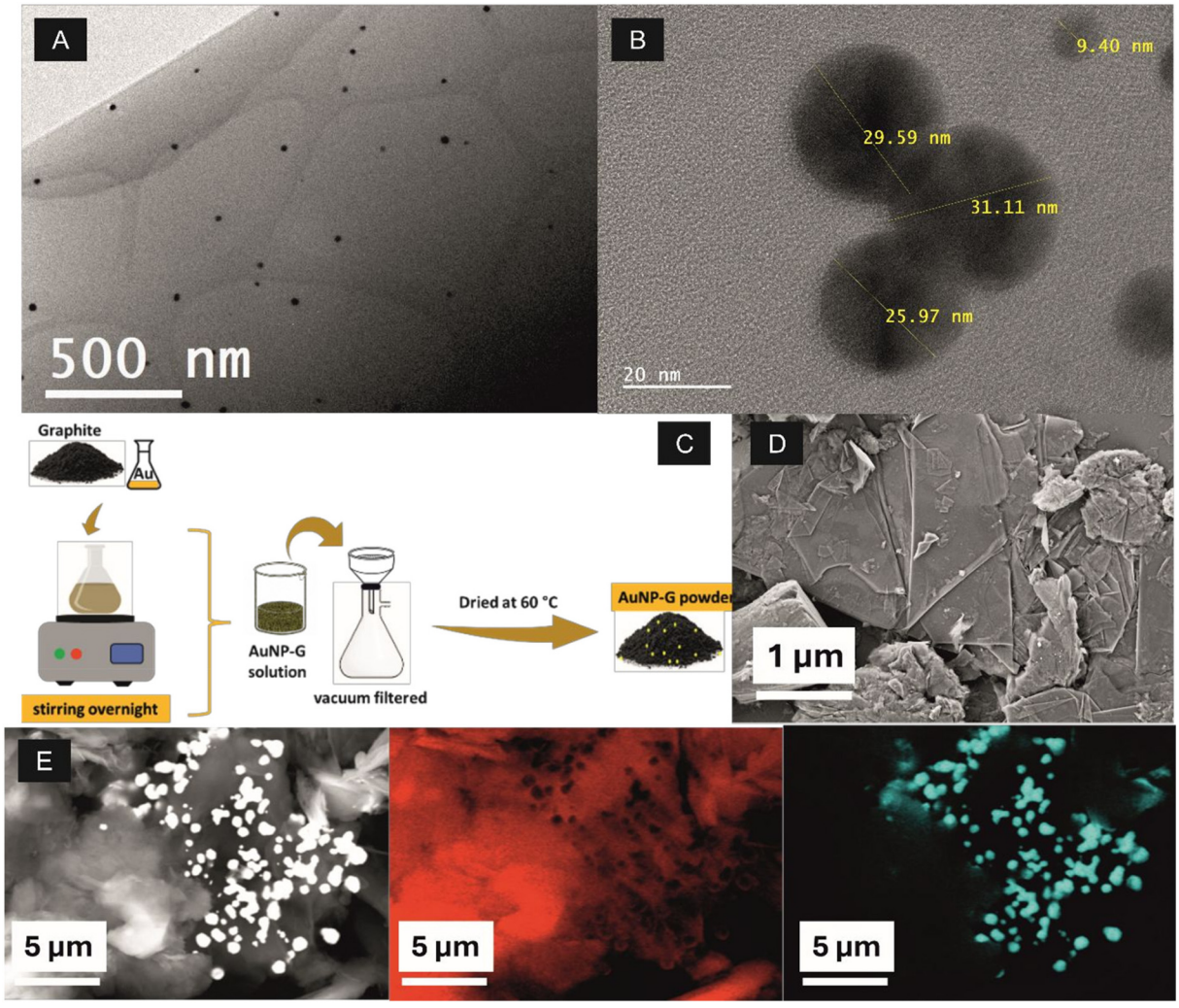

Fig. 4 (A) and (B) TEM images of AuNPs formed within castor oil. Reproduced with permission from ref. 93. Copyright Elsevier 2025, Creative Commons License CC BY 4.0. (C) Schematic of the synthesis of AuNPs on graphite flakes, (D) SEM image showing the synthesised AuNPs on graphite, and (E) EDX maps of the presence of AuNPs on graphite, which the overall image (left), carbon map (middle) and Au map (right). Reproduced with permission from ref. 94. Copyright Royal Society of Chemistry 2025, Creative Commons License CC BY 3.0.

coke, which is an extremely emission and energy intensive process due to the complexity of the supply chains involved.[89] With all of this in mind, improved methods of conductive carbon production should be sought, with sustainability and scalability at the forefront. One potential source is that of biochar, a carbon-rich material that is produced from the pyrolysis of biomass.[90] Recently, initial studies have reported the first use of biochar within conductive filament for additive manufacturing electrochemistry. These studies have used coconut waste[91] or softwood pellets[92] to produce biochar, which can then be incorporated alongside carbon black in the filament. These works have the effect to reduce the overall reliance on petroleum-based carbon sources, but more work needs to be completed in this area to truly realise its potential.

In addition to simply loading filament with carbon, recent work has looked to increase the functionality of the final filament through the inclusion of metallic nanoparticles at early stages in the additive manufacturing process. Two strategies have been reported for the integration of gold nanoparticles (AuNPs) with focusses on sustainability. Firstly, the synthesis of the AuNPs was reported within castor oil, Fig. 4A shows the TEM images, which was then utilised as the plasticiser within filament production.[93] The AuNPs were formed through the addition of KOH and raised temperatures, which reduced the Au(III) salts with the nucleation first starting in the aqueous phase before transferring to the oil phase. Even though this method creates a low loading of AuNPs within the castor oil, and the oil only makes up 10 wt% of the overall filament, a significant increase in the electrochemical performance was observed. To increase the metal loadings, a second method of forming AuNPs on graphite flakes has been reported, Fig. 4B.[94] In this work, the authors use the natural reducing ability of graphite to form the AuNPs. By immersing graphite flakes within an aqueous solution of Au(III) salts and stirring overnight, the graphite flakes become decorated with AuNPs. These AuNP modified graphite flakes were then incorporated into the filament, producing an enhanced performance toward the detection of Pb^{2+} within environmental waters. This method has since been shown for the production and incorporation of silver nanoparticles (AgNPs)[95] and platinum nanoparticles (PtNP).[96] These produced a platform capable of detecting Cd^{2+} well below World Health Organization targets in environmental waters,[95] and a platform capable of producing green hydrogen, respectively.[96] These examples show how improvements in material composition and performance can be achieved with sustainable approaches. With all these fillers it is important for future work to establish potential issues in regard to leeching from the filament upon printing or from electrodes upon use, which, depending on the filler, could release potentially toxic substances into the environment.

As seen above, there is a widening range of fillers that have already been incorporated into filament, but there is significant space for this to expand. This could be carbon-based materials such as activated carbon, MXene and carbon nanohorns, other metal and metal–oxide based fillers such as Nickel, ZnO, or TiO_2, or conductive polymers such as poly (pyrrole) (PPy), or poly(aniline) (PANI). We believe the extreme customisation available through additive manufacturing has excellent synergy with specialised functional fillers.

4. Design and printing

Once filament has been produced, the electroanalytical platform to be used can be designed and printed. The use of additive manufacturing within labs gives researchers the unique opportunity to rapidly prototype designs and allows them to generate whole sensor concepts. When considering the design of cells, the parts for the sensing platform can be printed separately and assembled post print Fig. 5A, or they can all be printed in a single print Fig. 5B.

The latter can be advantageous to save time, but consideration of the waste generated should be made, which can come in two main areas. Firstly, in the printing process, single-extruder printers can be used for multi-material prints but the previous filament must be purged from the nozzle on each change meaning large amounts of waste can be generated. As such, it is advantageous to use multi-extrusion/toolchanger printers, which can have filament loaded into different extruders and used when required. Secondly, when cross-contamination is an issue, each platform is essentially single-use, in particular for PLA-based devices due to the solution ingress.[55] As such, for development and testing it can be beneficial to use cells printed from a water-resistant material such as PETg or ABS, which can be easily cleaned and re-fitted with a new electrode, significantly reducing waste generation.[9,98]

When designing specific parts, there are parameters to consider that can improve the sustainability of your print, whilst also improving the performance. Firstly, ensuring the connection length from the working electrode to the potentiostat connections is as short as possible.[99] Filament is primarily made of insulating polymeric material, and as such any printed part is not a near-ideal conductor as used within classical electro-

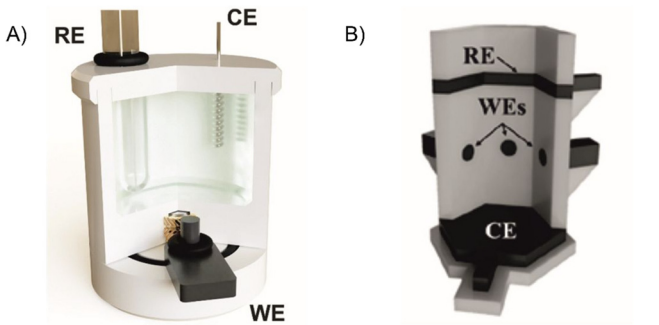

Fig. 5 (A) Cell created using separate pieces. Reproduced with permission from ref. 75, copyright Royal Society of Chemistry 2023, Creative Commons License CC BY-NC 3.0. (B) Cell printed all in one. Reproduced with permission from ref. 97, copyright American Chemical Society 2023, Creative Commons License CC BY 4.0. In both cases, RE = Reference Electrode, CE = Counter Electrode, and WE = Working Electrode.

des. Therefore, maintaining the shortest connection length between the electrode|solution interface will not only give the best electrochemical performance, but it will also reduce the amount of material used as well as reduce the overall cost of production. Next, within the design phase the user has control over how solid the part is by modifying the infill. A recent study has shown that for commercially purchased conductive PLA, the infill percentage could be reduced to only 30% rather than 100% which represents a solid electrode, without significant deterioration in the electrochemical performance.[100] This offers another way to reduce the amount of plastic used per part. It should be noted that this has only been tested on a single filament and how this translates to bespoke filament needs to be explored.

5. Post-processing

Within the literature of additive manufacturing electrochemistry, a process called activation is commonly used to increase the part performance. This is due to a gloving effect that occurs when extruding filled thermoplastics, with the polymer parts migrating to the surface. This means that the final printed parts tend to have a thin, insulating polymeric covering at the electrode|solution interface. This is no issue when exploring outer-sphere redox probes, which can complete electron transfer processes through quantum tunnelling; however, when studying molecules that use an inner-sphere mechanism, the removal of this layer is vital to achieve the best possible performance. Fig. 6 illustrates differences in scanning electron micrographs for an electrode printed from conductive PLA before and after an activation process.

It is important to note that the activation of commercially available filament is essential to achieve any real electrochemical response, however for bespoke filaments good signals can be obtained without activation, but it has been shown to still improve the performance.[57,95] Therefore, whether or not to activate a bespoke electrode is dependent on the performance requirements for any given application. Due to the field of additive manufacturing electrochemistry mainly functioning around commercially available filaments for many years, there has been a plethora of activation methods reported and compared for their performance.[101] These include simple mechanical polishing, electrochemical activation within buffer solutions, acids, or bases, soaking parts within solvents such as dimethyl formamide or acetone, carbonisation of the surface, thermal annealing using reducing agents such as ascorbic acid, or a combination of these.[102–109] When considering which to choose, clearly the best option is none if your electrode can meet the required levels for the application. But if required, avoiding the use of harsh solvents or high temperatures is preferable, with good results seen from simple mechanical polishing or electrochemical activation within standard aqueous based solutions. Although it must be noted that the impact of microplastic generation through mechanical polishing of additive manufactured electrodes has yet to be explored.

6. End-of-life considerations

We have discussed many ways in which the process of creating electroanalytical platforms through additive manufacturing can be improved in terms of sustainability, but dealing with the items after their working life has finished remains vital. If electroanalytical platforms are printed as separate parts, it makes recycling of the material much simpler. A recycled PLA-based cell has been shown to be cycled up to four times back into filament to reprint the same cell.[57] In a similar way, ABS filament has been shown to be recycled to print a mould for paste deposition and electrode production[110] or used as a filler in the paste itself.[111] It has also been shown that non-conductive PETg prints have been used as the base polymer for the production of bespoke conductive filament.[42,43] These systems show promise in making use of spent material, but there will always be a limit on the amount of times the material can be

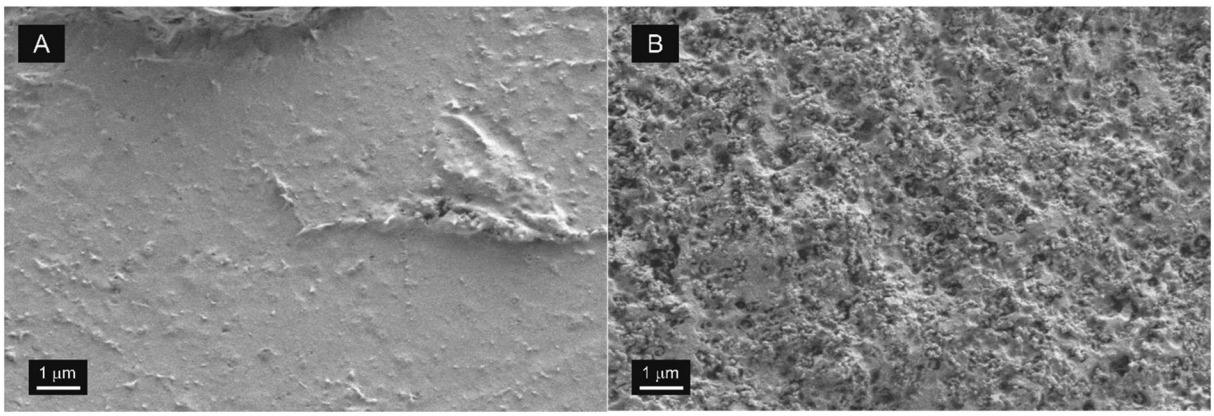

Fig. 6 (A) SEM image of an electrode surface printed from a carbon black/PLA bespoke conductive filament. (B) SEM image of an electrode after electrochemical activation within 0.5 M NaOH. Reproduced with permission from ref. 75, copyright Royal Society of Chemistry 2023, Creative Commons License CC BY-NC 3.0.

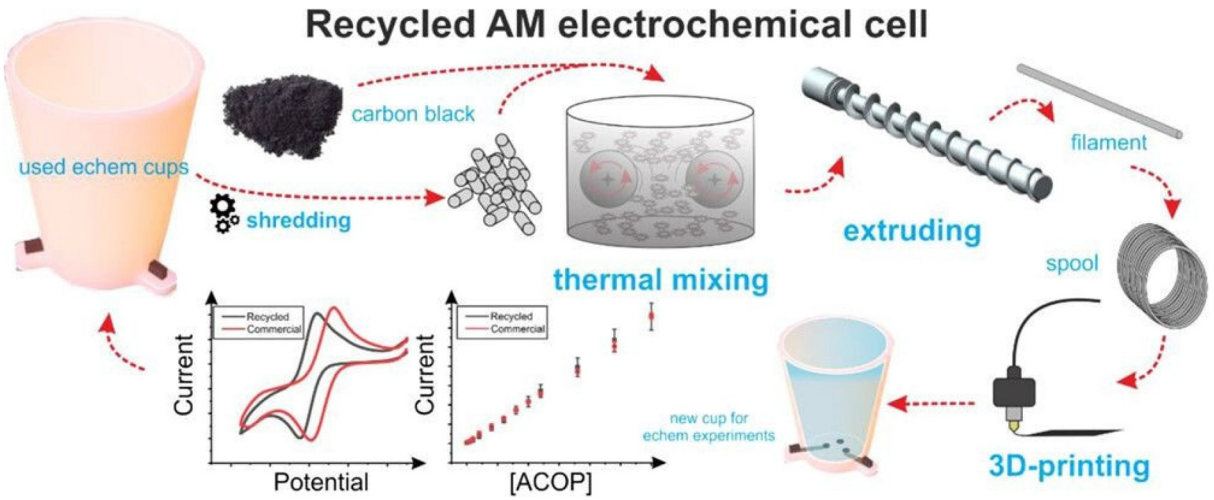

Fig. 7 Schematic highlighting the recycling of a mixed material electroanalytical sensing platform back into filament to reprint an identical cell for the same application. Image reused with permission from ref. 113, copyright American Chemical Society 2023, Creative Commons License CC BY 4.0.

processed before it becomes unusable due to the reduction in polymer chain lengths through mechanical recycling.[112] Potentially, at this stage the gradual incorporation of low-quality polymer within a larger sample of high quality polymer is a way to continue to reuse this but this needs to be explored.

In terms of mixed material prints, work has been reported on the recycling of full electroanalytical cells, Fig. 7.[113] In this study the authors explored four processing techniques (granulation, ball-milling, solvent mixing, and thermal mixing) to return the cells back into filament. Using another thermal mixing step to disperse the concentrated areas of conductive filament, the authors were able to produce both conductive and non-conductive filaments. The non-conductive filaments were possible as the electrodes in the electrochemical cell comprised of such a small percentage of the overall design there was not enough carbon black within the recycled filament to induce conductivity. To create the conductive filament, additional carbon black was added to the thermal mixing step. Through this method, a new fully recycled electrochemical sensing platform was printed and shown to offer the same electroanalytical performance for the detection of Acetaminophen as achieved with the original. This can be seen as an important piece of work not just for full cells, but also purge material, which FFF produces a significant amount of, with complete purging between filaments being required to ensure good quality prints and not short-circuiting. This can create a considerable amount of lab waste. Although showing a unique way to recycle these mixed material prints, increased amounts of thermal processing will speed up the polymer degradation and limit the number of times it can be recycled. As such, a preference is placed on printing individual parts, especially within research environments where multiple experiments are needed to be performed.

For using within electroanalytical applications, only mechanical recycling techniques such as these have been explored. Mechanical recycling is a simple and widely available methodology, applicable to in-house recycling programmes; however, there are considerable drawbacks due to issues with contamination and the reduction of polymer chain length with every reprocess, hindering the mechanical properties of the polymer and limiting its use within future filament.

An alternative is chemical recycling of the polymers,[114–116] however this has yet to be reported in the context of additive manufacturing electrochemistry. Even so, this should be an area explored in the future. Instead of physically reprocessing the waste as in mechanical recycling, chemical recycling looks to break down the polymer chains into their monomeric or oligomeric building blocks through depolymerisation, pyrolysis or solvolysis. These recovered feedstocks can then be purified and repolymerised to produce virgin-quality filaments. This can potentially enable closed-loop recycling, even for contaminated and degraded prints, maintaining mechanical integrity. It is also important to note that the majority of published works have focussed only on recycling PLA-based devices. Now that some procedures for this have been established, we expect to see these applied to a wider range of polymers used within the field, such as PETg, PP, and TPU.

7. Sustainability analysis

7.1. Life cycle analysis/assessment (LCA)

Life cycle analysis (LCA) is an important tool that is now widely used. It is a systematic method for evaluating the environmental impacts associated with all stages of a product's life, incorporating the raw material extraction, manufacturing, use, and the end-of-life disposal or recycling. When considering additive manufacturing, LCA can provide critical insights into the differences in sustainability between processes by quantifying energy consumption, material efficiency

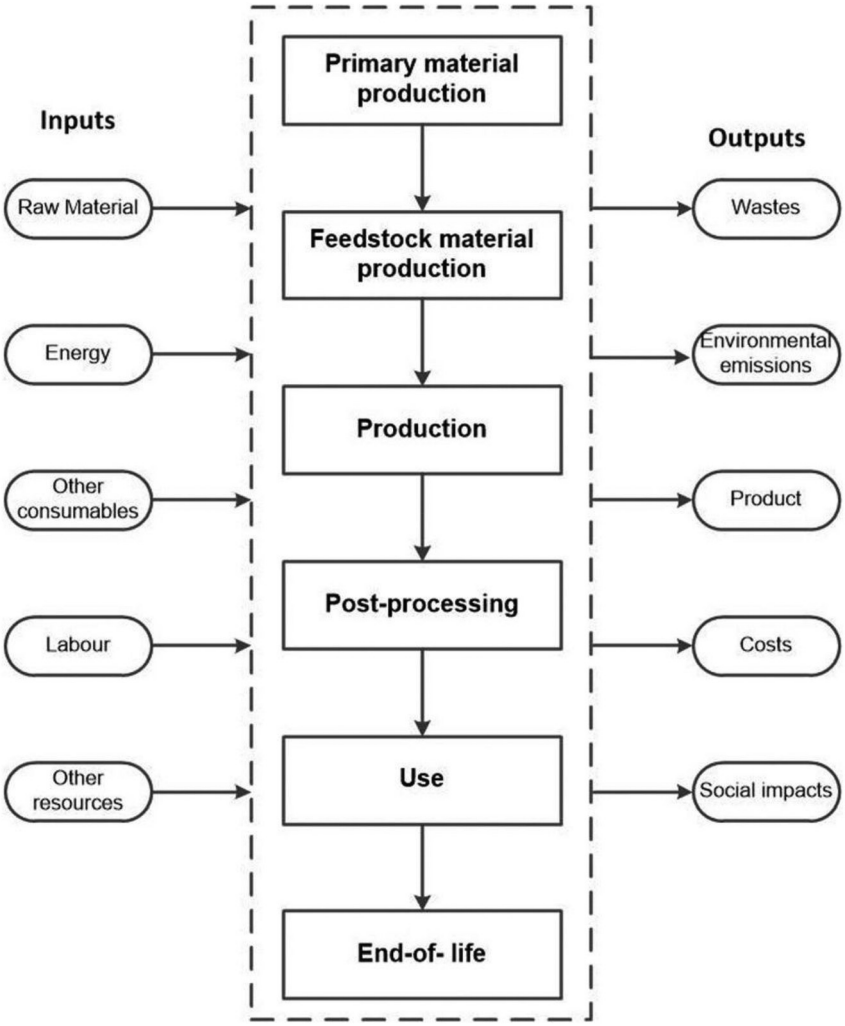

Fig. 8 A life cycle of an additively manufactured part. Reproduced from ref. 118. Copyright Elsevier 2023, Creative Commons License CC-BY.

emissions, and waste generation.[117,118] A schematic explaining considerations in LCA for additive manufacturing can be seen in Fig. 8.

One can see the scale of considerations that must be taken into account when performing a LCA, and we suggest that the field would benefit from future work integrating this into their reasoning. Many output areas are often not considered with FFF printing. Due to its synonymity with home and hobby printing, one such area is the emissions, which can generally come in two forms, Volatile Organic Compounds (VOC) and Ultrafine Particles (UFP) emissions. For UFPs it has been shown that with typical FFF printing, particles typically ranging from 20–100 nm are regularly detected, with studies now showing particles below 4 nm have been shown to be emitted.[119] With FFF printing, these emissions typically occur at rates of 10^7 to 10^{11} particles per minute, depending on the filament type, printer model and printing temperature.[120] These UPF's can enter the body through the lungs and translocate to essentially all organs, and have been shown to cause more pulmonary inflammation than larger particles.[121] In terms of VOC's, this is highly dependent on the material being printed. For example, when printing ABS it has been shown that styrene is emitted, which is classed as a probable carcinogen to humans. In comparison PETg has been shown to emit predominantly acetaldehyde and has been shown to be a lower emitting filament.[122]

It is important to understand these studies are for traditional FFF materials and currently do not consider filaments highly loaded with fillers. For these reasons it is highly recommended that all FFF based labs ensure the presence of HEPA filters (H12/H13) and/or near-source extraction when printing until more research and data is available on these crucial topics.

7.2. AGREE metrics

With sustainability becoming an increasingly central concern within analytical science, more specific metrics for the field have emerged as essential tools for evaluating the environmental impact of laboratory methods.[123–127] One such method is the Analytical GREEnness (AGREE) metric,[128–130] which

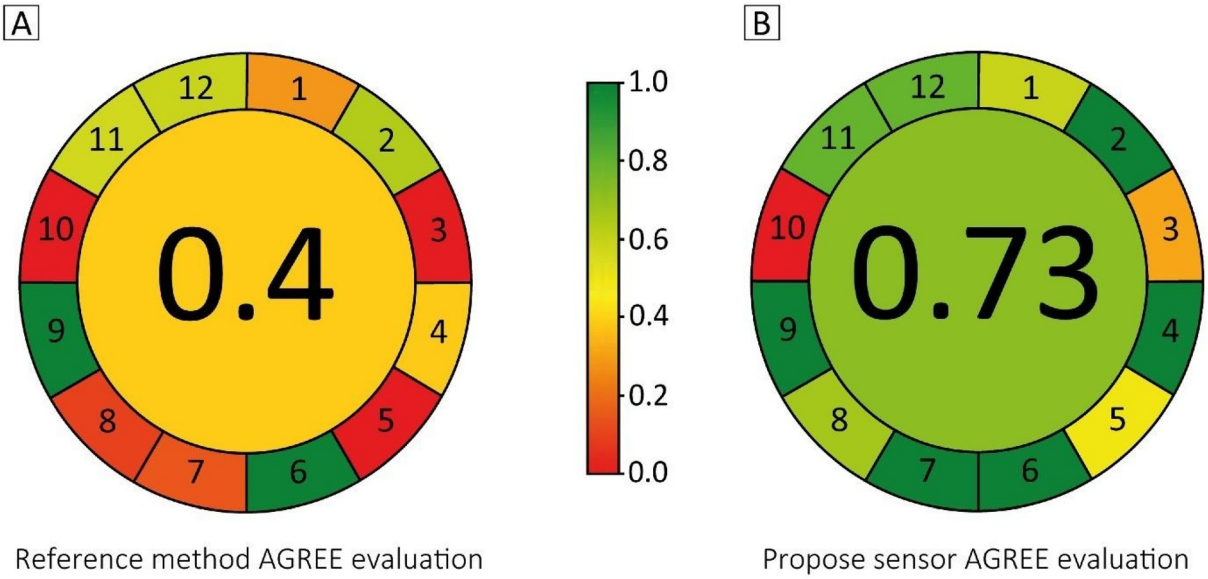

Fig. 9 Example evaluation of the AGREE metrics, comparing a reference method (A) and a newly proposed methodology (B). Figure reproduced from ref. 134. Copyright Elsevier 2025, Creative Commons License CC BY NC ND.

offers a comprehensive framework for assessing how well an analytical procedure aligns with the 12 principles of green analytical chemistry.[131] AGREE enables researchers to make informed decisions about method development and optimisation. The 12 principles it considers are: (1) direct analytical techniques should be applied to avoid sample treatment; (2) minimal sample size and minimal number of samples; (3) *in situ* measurements should be performed; (4) integration of analytical processes and operations to save energy and reduce reagent use; (5) automated and miniaturised methods should be used; (6) derivatisation should be avoided; (7) generation of large volumes of analytical waste should be avoided and proper management of waste should be provided; (8) multianalyte or multiparameter techniques are preferred; (9) energy use should be minimised; (10) reagents obtained from renewable sources are preferred; (11) toxic reagents should be eliminated or replaced; (12) safety of operator should be increased.[129]

For electroanalysis, these metrics encourage the development of techniques that minimise resource use, reduce hazardous waste, and improve energy efficiency, all while maintaining analytical performance.[132,133] This is particularly relevant for FFF-based electroanalysis, which aligns well with these principles through its ability to enable prototyping of compact electrodes with ultra-low waste generation. The synergy between FFF and electrochemistry allows for the use of direct analytical techniques by integrating the sensing platforms and printed devices, often eliminating the need for extensive sample preparation. Moreover, the use of recycled material (as discussed earlier) further supports minimal waste generation. An example of how these metrics look is shown in (Fig. 9), where Mazzaracchio *et al.*[134] explore the development of a greener methodology for the detection of phosphate, comparing a classical reference method to their new proposal using additive manufacturing. Although not an electrochemical method, the comparison between a reference and the new method is a good way to contextualise the improvements. We suggest the adoption of this.

There are also examples of electrochemical sensors utilising AGREE,[134] and we suggest that FFF-based electroanalytical devices can move toward the use of this metric.

8. Conclusions and future outlook

Within the field of additive manufacturing electroanalysis there has been significant strides toward improving the sustainability of bespoke filament production with excellent examples transitioning toward utilising recycled polymers alongside more sustainable additives. Additionally, a key advancement has been the development of conductive filaments with different base polymers that improve the chemical and electrochemical stability of the electrodes, avoiding single-use electrodes. Although some studies have been published on recycling of electrodes or devices, more work is required considering the end-of-life processing of electrodes and electroanalytical platforms. Inherently, electrodes will always have limited use due to the requirement of avoiding cross-contamination, and therefore some of the biggest gains in sustainability will be found through end-of-life considerations and the movements to a true circular economy. It is noted that the vast majority of work improving the sustainability of filaments within this field is dedicated to the use of recycled polymers. Although admirable, when considering the "3 R's" of Reduce, Reuse, and Recycle, recycling is the least favourable option. More work should be dedicated to the reuse of systems when employing more stable base polymers, as well as the reduction

of certain components. One example of this has been reported recently, replacing 10 wt% of PLA with microcrystalline cellulose.[135] This strategy produced significant improvements on commercial filament with the same carbon loading and we expect further strategies such as this to be explored.

Conflicts of interest

There are no conflicts of interest to declare.

Data availability

No new data were created or analysed during this study. Data sharing is not applicable to this article.

Acknowledgements

We would also like to thank the EPSRC (EP/W033224/1), RSC Sustainable Laboratories Grant (L24-3644795451) and Horizon Europe (grant 101137990) for funding.

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