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Additive manufacturing for electrochemical labs: An overview and tutorial note on the production of cells, electrodes and accessories

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ABSTRACT

Additive Manufacturing (AM) is an ever-growing part of modern scientific research due to its ability to create complex features, low wastage, ever-decreasing cost of entry and rapid prototyping capabilities. Up to this point, the use of AM in electrochemical research has focused around two of the main components of the experimental setup: the working electrode, and the electrochemical cell. In this paper we highlight how researchers have utilised AM in the literature and offer our own insights into how this technology can be exploited to benefit all areas of electrochemical research. For the development of electrodes, much of the literature utilises commercially available conductive PLA filaments in conjunction with FFF printing, with only a few groups expanding into the development of their own bespoke conductive materials.

AM offers huge advantages in the production of electrochemical cells, allowing users to produce bespoke designs to fit their experimental needs, rapidly producing these at low cost and easily modifying the design to improve performance. However, the use of AM in electrochemical laboratories should not stop there. We present basic designs of electrodes, cells and even accessories that can benefit all electrochemical researchers (new and experienced) in their quest for reproducible and reliable results. These designs are offered free of charge, are available to download from the Supporting Information and can be easily modified to meet any users' specific needs. As such, we feel AM should be a staple of every laboratory and hope this work inspires people to think about all the ways that AM can benefit their research environments.

Introduction: Additive manufacturing and electrochemistry

Additive manufacturing (AM, 3D-printing) has become an indispensable part of modern-day life, transcending multiple industrial categories, with research spanning a variety of broad fields and increasingly becoming a household staple. Although the cost of certain printers remains extremely high (with some concrete 3D printers being sold for prices in the £millions), robust and reliable printers can be purchased easily for under £500, allowing AM to be increasingly accessible to the individual and research groups who are not focussed solely on the area. AM allows for 'bottom up' manufacturing, providing the ability to construct items in a layer-by-layer approach. Since the first invention of stereolithography (SLA) in 1983 by Charles "Chuck" Hull [1], the field has rapidly expanded to incorporate various printing methodologies -selective laser sintering (SLS), fused filament fabrication (FFF) and direct metal laser sintering (DMLS) to name a few- that are capable of printing a vast array of materials (plastic, metal, ceramic, concrete etc) [2]. The process of AM begins with the creation of a CAD (Computer Aided Design) model of the item to be fabricated/printed, which is then converted to an STL (Stereolithography) file. This file is then processed in a Slicer Software to produce a G-code file, which can then be 3D printed/ additively manufactured by the device in sequential 2D layers. There is a plethora of software available for completing these tasks, in the work presented by us Fusion 360 is used for the design of parts. This is a cloud-basedCAD software that is free to use for non-commercial users, making it more accessible to low-budget users than some more traditional CAD packages. For slicing the G-code, most printers have specific software however, our work uses Prusa Slic3r which is a free, open-source slicing software based on the Slic3r open-source toolbox (a community driven project in the RepRap community). The direct translation of design to component, no requirements for additional tooling, ability to create complex internal features, rapid

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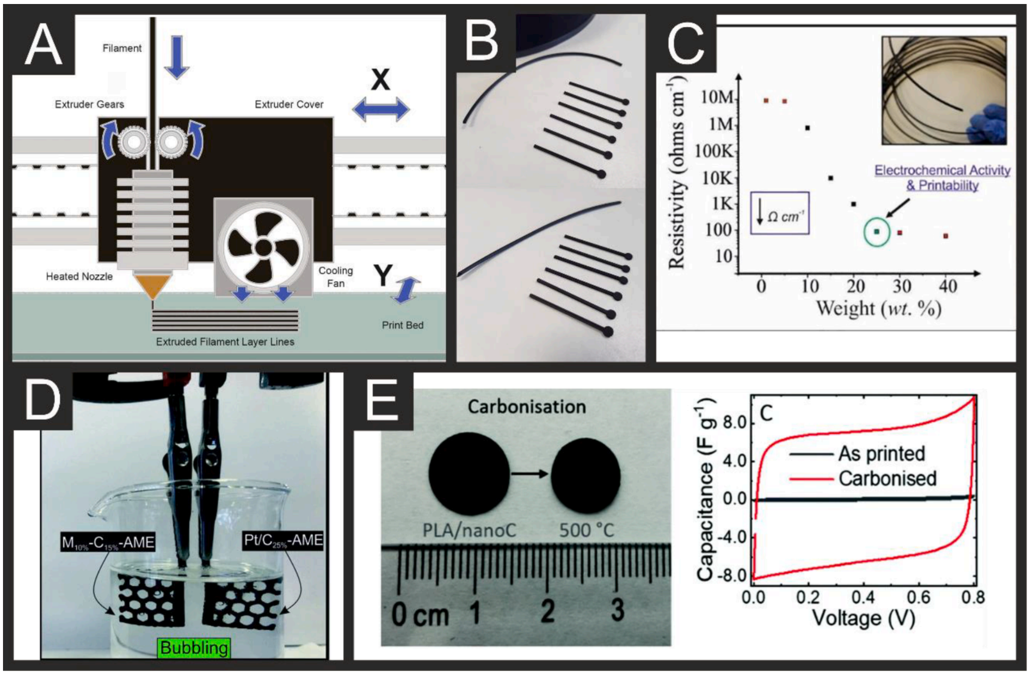

Fig. 1. A) Schematic of the process of FFF printing. B) Examples of commercial conductive PLA filaments Black Magic (top) and Protopasta (bottom) and some example working electrodes printed with them. C) Resistivity *vs.* NG content of different working electrodes printed using bespoke filaments. Reproduced with permission from ref [27]. Copyright 2020 Elsevier. D) Image of water splitting enabled utilising a $M_{10\%}C_{15\%}$-AME and a Pt/$C_{25\%}$-AME as the cathode and anode, respectively. Solution composition: 0.5 M H_2SO_4. Cell voltage applied using a commercially available alkaline zinc-manganese dioxide 1.5 V battery. Reproduced with permission from ref [30]. Copyright 2019 Royal Society of Chemistry. E) Photograph of the 3D-printed PLA/nanoC electrodes before and after carbonisation (left). Capacitive behaviour of the 3D-printed electrodes by means of cyclic voltammetry before and after thermal activation at 500°C. Reproduced with permission from ref [42]. Copyright 2020 Royal Society of Chemistry.

prototyping and extremely low (often zero) wastage, among other advantages, have all contributed to the expanding popularity of AM [3]. All of this in conjunction with the continually falling costs of entry into AM has allowed the industry to rapidly grow.

As expected with such a versatile and rapid technique, research into AM spanning a vast array of topics has been reported, such as mechanical engineering [4], aerospace design [5], medical research [6–8], sensors [9–11] and its role in sustainability [12]. The future of AM for the production of 3 and 4-D printed materials in research has also been summarized [13]. AM has also been applied to electrochemistry [14] with the research predominantly focused on the development and use of conductive materials for applications in the fields of energy storage [15-–17] and sensors [18–20]. Additionally, there is a large amount of work currently being done to improve the sustainability of AM, through the recycling of used filaments or transformation of other plastics into printable filaments [21,22]. The authors note that each new filament should ideally be calibrated on your printer and suggest printing a temperature tower with each new material to easily determine ideal nozzle temperatures for optimal printing of each new material. The rapid prototyping capability of AM had allowed researchers to produce components/wearables for these devices *in-situ* to test their capabilities, saving time and resources. However, this technology should not be observed as only for researchers dedicated to the area of AM, but rather it can be seen as a vital tool for improving the research quality of all electrochemical labs. Thompson and Agrawaal [23] have recently overviewed the application of AM applications in the field of analytical chemists, concluding that it has a useful place in laboratories due to its simplicity and ability to rapidly produce analytical devices on-the-spot, at low cost, completed in single-steps and made with complex geometries from multiple materials in a matter of hours. Consequently, in this manuscript, we will present a tutorial overview of the uses of AM in current electrochemical research, and how to accommodate AM into the users own lab through the rapid and cost-effective fabrication of electrochemical laboratory devices. From electrodes and electrochemical cells to general accessories used in the lab, we include explanations of why they are useful and provide the appropriate STL files for users to download and print “as-is”, while allowing the ability to rapidly modify and adapt to their own experimental/requirements/set-ups. In this work we use two 3D-printers, a Prusa i3 MK3S+ (Prusa, Czech Republic) and a Raise3D E2 (Raise3D, US), although there has been a published review on selecting 3D printers for labs [24]. Note that we recommend and encourage individuals to produce these items in recycled filament wherever possible.

Additive manufacturing of electrodes

The working electrode is one of the most significant parts of the electrochemical set-up, it possesses the interface of interest [25]. As such, a vast majority of the literature using AM in electrochemical research is focused on the optimization of working electrodes or the development of new materials for them. A summary of the literature found on this area is provided in Table 1, where a significant shift has taken place from the use of Selective Laser Melting (SLM) to fused filament fabrication (FFF) for the production of working electrodes. It is this shift to FFF, Fig. 1A, that has opened the field of AM to a far wider audience, with excellent FFF printers available for a few hundred pounds, and some can even be purchased for under £100. As FFF uses a solid filament, that is extruded and then printed directly onto the print bed, there is also minimal waste and minimal post production making it highly suited to a research environment. From inspection of Table 1, it is noticeable that the most commonly used filaments in the production of working electrodes are polylactic acid (PLA) based, that is normally impregnated with a carbon based nanomaterial to produce a conductive material. There are two frequently used PLA/carbon filaments that are commercially available; Black Magic 3D which is a graphene based PLA filament which has a quoted filament volume resistivity of 0.6 Ω cm^{-1} and Protopasta Conductive PLA which utilises carbon black and has a quoted filament volume resistivity of 15 Ω cm^{-1}, Fig. 1B. There are increasing amounts of alternative conductive filaments available for commercial purchase, such as Electrifi (Multi3D, US), SUNLU conductive ABS (SUNLU, US), Amolen conductive PLA (Shenzhen Amolen Technology Development Co., China) and 3dkonductive PLA (3dk Trading GmbH, Germany) to name a few, however these rarely if ever have featured in the literature. PLA has become a staple of the field as it possesses a wide printing temperature window, facile mixing and uniform polymer chains, giving it a forgiving printing process, even with significant additives. These advantages currently outweigh the negatives of not being waterproof, poor strength, hydrolytic and photolytic

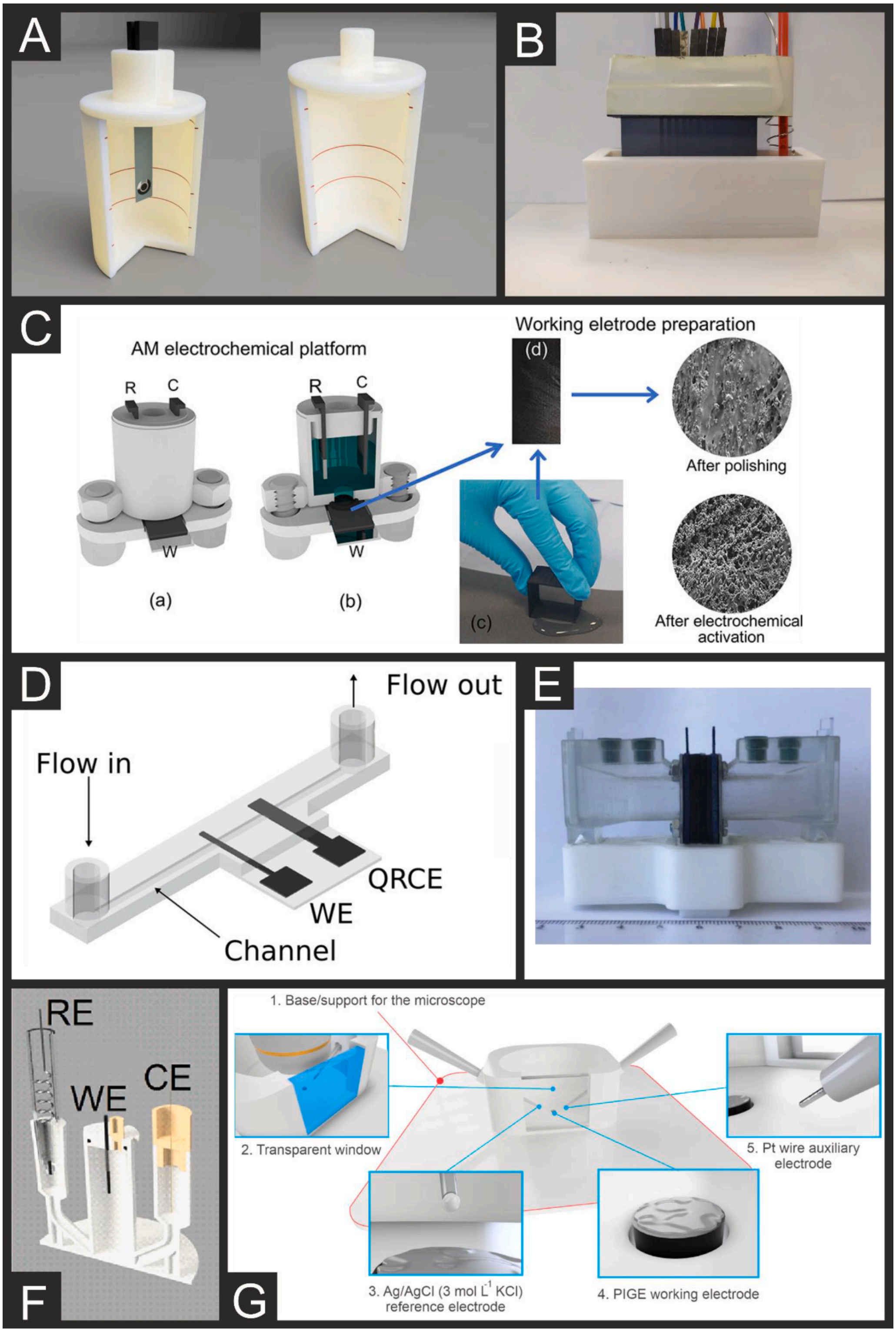

Fig. 2. A) Fusion 360 renders of the volumetrically graduated electrochemical cells with an SPE holder lid (left) and a standard electrode holder lid (right). B) Photograph of the small AM box cell (volume = 10 mL) produced for electrochemical experiments with array SPE electrodes. C) Schematic diagrams of the AM electrochemical platform and the 3D-printing working electrode preparation procedure: (a) complete AM electrochemical platform; (b) transversal cut view of the AM electrochemical platform (total volume = 5 mL); (c) polishing procedure of the hollow printed cube (3.0 cm × 3.0 cm × 1.5 cm and thickness of 0.75 mm); (d) rectangular printed base after polishing (3.0 cm × 1.5 cm and thickness of 0.70 mm). Other information: (R) 3D-printed pseudo-reference electrode; (C) 3D-printed counter electrode; (W) 3D-printed working electrode. SEM images of the polishing 3D-printed working electrode before and after electrochemical activation are also shown. Reproduced with permission from ref [39]. Copyright 2019 American Chemical Society. D) CAD drawing of a channel flow cell with a 1.5 × 1.0 mm channel and two band electrodes. Reproduced with permission from ref [52]. Copyright 2019 Elsevier. E) Photograph of the 3D-printed electrochemical water splitting cell assembly, with bottom white support to hold the cell. Reproduced with permission from ref [55]. Copyright 2019 Wiley. F) Cross-sectional CAD drawing of the integrated electrochemical platform for electrochemistry. Reproduced with permission from ref [58]. Copyright 2020 Elsevier. G) Electrochemical cell designed and 3D-printed using FFF for coupling of VIMP and Raman in-situ. Reproduced with permission from ref [60]. Copyright 2021 Elsevier.

degradation and slightly increased average cost compared to other commonly used filaments such as acrylonitrile butadiene styrene (ABS) and Polyethylene Terephthalate (PET).

Currently there has been few reports on the development of bespoke filaments, produced by research groups specific for electrochemical applications, such as PLA modified with nanographite (PLA/NG) for electroanalytical sensing [26–28], $LiFePO_4$ and carbon nanotubes (LIF/CNT) for Li ion batteries [29] and 2D-$MoSe_2$ with Super P for hydrogen and oxygen evolution applications [30]. The initial work by Foster *et al.* [27] to produce their bespoke PLA/NG filament managed to increase the amount of active component in the PLA from ~8 wt%, which is found in commercial filaments, up to 40 wt%. The authors found the optimum composition for the best compromise between electro-analytical and printing performance was at 25 wt%. Below 20 wt % did not meet the required percolation threshold and above 25 wt% the lack of thermoplastic binding material resulted in a brittle, non-reproducible print, Fig. 1C. Hughes *et al.* [30] took the this idea further through incorporating both a carbon based nanomaterial (Super P) and the transition metal di-chalcogenide 2D-$MoSe_2$ to produce a working electrode suitable for the Hydrogen Evolution Reaction (HER). This again incorporated 25 wt% additive into the PLA matrix, with 10 wt % 2D-$MoSe_2$ and 15 wt% Super P. In this work, the authors also investigated one of the other main areas of research in this field, changing the physical parameters of the electrode. AM lends itself extremely favorably to the rapid production of novel electrode shapes and dimensions, requiring small modifications in the STL and G-Code files which can be completed with the print restarted in minutes. The authors achieved this through the development of "flag-style" electrodes, Fig. 1D, tailored to produce a high surface area electrode and enhanced performance for water splitting. As can be seen from Table 1, there have been numerous shapes produced for AM electrodes reported using vastly different dimensions such as helices [31], bands [32], coins [33], fractals [34] and most commonly discs [35]. Observed throughout the literature is a huge variety of disc electrodes of varying dimensions and little to no discussion of the reasoning behind the choice. We direct readers to the Supporting Information Table 1, where an STL file is available for a parametric disc or "lollipop" electrode in a set size of 3 mm diameter disc and 1 mm thickness connected to a stem of length 20 mm and width of 1 mm, which can be modified easily in CAD software to the required dimensions. Typically, these will be printed in some way to mimic traditional electrodes such as glassy carbon, screen-printed or gold electrodes, with developments toward determining the relationship between the geometrical area and electro-active area of these AM electrodes [36]. The field of AM electrodes, although expanding, is still in its infancy with the majority of reports still utilising the same commercial filaments with approaches to improve their performance through "activation" of the electrodes. In short, this typically is achieved by effectively removing some of the PLA matrix after the printing of the electrode to make more of the conductive additive accessible for electrochemical studies. Various activation methodologies have been reported such as simple mechanical polishing [37], electrochemical scanning in PBS [26], acid [38] or NaOH [39,40], soaking in solvents such as DMF, acetone, ethanol or methanol [41], carbonisation [42], thermal annealing [43] or more recently through the use of reagent-less laser treatment [44] and "greener" reducing agents [45], Fig. 1E. Although there is still work to be do in this aspect, the optimization of the electrochemical performance of AM electrodes needs to be achieved through all areas of their production. We feel that the biggest steps to achieve the best performances is through the generation of novel filaments, be that the development of new additives or the incorporation into different polymer matrices. However, as can be seen throughout the literature highlighted in Table 1, significant strides have been made in the development of electrochemical systems utilising commercially available filaments on low-cost 3D-printers. This demonstrates that this area of work is not only accessible to the "ultra-rich" research groups anymore. Furthermore, there are significant advantages to having 3D-printers available in the lab for other aspects of work, such as quick prototyping and the development of electrochemical cells, which we will discuss next.

Additive manufacturing of electrochemical cells

Electrochemical cells can come in all shapes and sizes depending on the required application of the research. For a lot of standardized electrochemical experiments, a three-electrode set-up is used, for which there are a plethora of common options, such as expensive glassware, vials, beakers and even shot glasses. Obviously, these vessels work perfectly fine for experiments that are repeated using standardized electrodes and common solution volumes by the experienced researcher. However, setting up electrochemical measurements for those new to the area can be fiddly, with small mistakes leading to short circuiting. As such, we have developed facile electrochemical cells printed from PETG filament along with lids that contain fixed slots for both standard sized electrodes, Screen-Printed Electrodes (SPEs) and thermometers to allow any user to maintain set distances between their electrodes and constantly monitor the temperature of the solution. We have also included a blank lid design for the researchers own modification. In addition to this, for researchers with dual printing capabilities there are options to include graduated volumetric fill lines for 10, 25 and 50 mL solutions, Fig. 2A. Smaller and larger cell volumes are easily manageable through simple modifications of the CAD files. These cells are similar in design to some examples in the literature [46,47] using a cylindrical cell, but have been designed specifically for standard 3 electrode electrochemical experiments where standard commercial electrodes or SPEs can be used immediately, or any other electrodes can be used with the simple modifications of the CAD files. We chose to use PETG for the development of these cells due to the fact it is non-hygroscopic, is food and skin safe, and is regularly used in medical and food applications due to chemical and radiation sterilisation methods not affecting it [48]. In addition, it possesses excellent mechanical properties, chemical resistance to acid and alkali and ease of printing. The vast majority, if not all desktop printers should be able to print PETG without alterations. We direct the reader to the Supporting Information Table 1 to download the appropriate STL files ready to print at home or in the lab. These electrochemical cells are extremely simple and low-cost to produce, as the cell with in-built SPE connection in the lid only costs £1.13 to print (£3.41 including the cost of edge connectors and leads), highlighting the beneficial nature of having 3D printers available in the lab. This practicality and rapid prototyping is further exemplified through the white ABS cell shown in Fig. 2B. Rather than using oversized glassware with large volumes of solution for custom produced array electrodes, a simple cell with a fill volume of 10 mL can be quickly designed, sliced and printed to help make electrochemical experiments simpler. It is the rapid prototyping and flexible design process that has come through as the main advantage to creating electrochemical cells in the literature. This started with simple enclosed structures to house the solution under investigation, typically designed for electro-analysis such as the detection of mercury [49] or for housing SPEs and providing reliable electrode coverage for the detection of bacteria [50]. The system devised by Siller *et al.* [50] utilised two different AM top plates depending on whether only the working electrode or the whole three electrode set-up needed to be covered in solution, with the plates being held in place through magnets and sealed with an O-ring. Other designs for electro-analytical cells have gone down similar routes with O-rings key for sealing the system, such as Richter *et al.* [39], Fig. 2C, who developed a fully AM electrochemical cell and electrode set-up. Similar to this, Cardoso *et al.* [51] also used AM screws to connect a baseplate and the main cell. In this set up the working electrode was located at the base of the cell allowing for adaptation of the cell between batch and flow electrochemical experiments. A similar design is reported by Elbardisy *et al.* [26], who reported an AM wall-jet flow cell for a electro-analytical sensing platform combining high performance liquid chromatography and electrochemical detection. A good comparison to these screw-sealed designs for bulk analysis that highlights the benefits of AM for electrochemical cell design is presented by O'Neil *et al.* [52], who developed a flow cell utilising a channel, Fig. 2D, of only 1.5 × 1.0 mm that could be printed in ~45 min. This cell has a printed inlet and outlet that allowed the solution to flow across the electrode surface for use in hydrodynamic electrochemistry. The majority of these examples utilised FFF printing for both the production of the electrodes and electrochemical cell. Alternatively, there have been reports in the field of water splitting on utilising SLA for a scanning flow cell [53] or SLM printing for the production of electrodes and then FFF for the development of the cell [54]. The approach using SLA printing allowed for the production of a transparent cell, which allowed the researchers to monitor the formation of gas much more easily and could be applicable to monitoring colour changes too. This approach for the development of a water splitting cell was taken further by Lee *et al.* [55], Fig. 2E, who utilise SLM for the

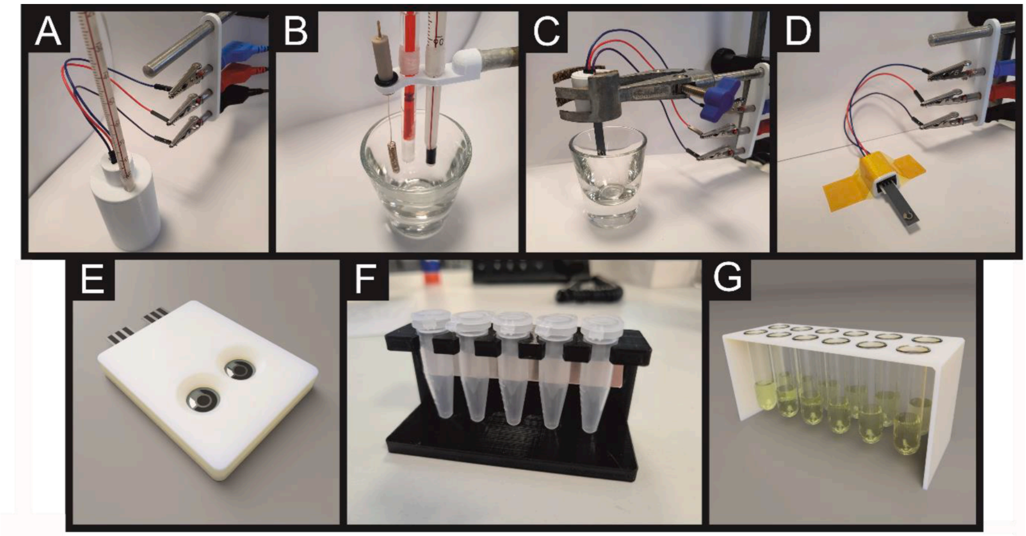

Fig. 3. A) Photograph of the AM clip holder and electrochemical flow cell with in-built SPE holder and thermometer holder. B) Photograph of a clamp holder for three electrodes/thermometer. C) Photograph of an SPE holder designed for use in a clamp. D) Photograph of an SPE holder designed for use in drop-casting or droplet experiments. E) Mask for use in drop-casting/functionalisation experiments on SPES. F) Render image of an AM small centrifuge tube holder. G) Render image of an AM test tube rack, which can easily be tuned to a centrifuge tube holder.

production of Ti and Ni based electrodes, FFF for the production of an ABS facile baseplate and SLA for the production of the translucent anodic and cathodic chambers. Some alternative uses for these electrochemical cells that have been reported include the conversion of magnetic nanoparticles [56], synthesis of magnetite nanoparticles [57] and the investigation and monitoring of the chemical reaction between carbon dioxide and hydrazine [58]. This latter example utilised the flexibility of AM to produce individual compartments for each electrode, Fig. 2F, which allow the reactants to be introduced to the system under oxygen-free conditions (Table 2).

Final examples of the benefits of AM for the production of cells are reported by dos Santos *et al.* [59] who produced a low-cost spectroelectrochemical cell for *in-situ* Raman measurements and da Silveira *et al.* [60] who have used FFF to generate a cell that allows them to combine electrochemical measurements and Raman spectroscopy *in situ*, Fig. 2G. The cell was made out of ABS which is more suited to the development of watertight cells than PLA, printed in a single-step with no assembly required and had a material cost of approximately £1.50. This is an excellent example of how AM can solve the challenges of performing multiple techniques at once, leading to more reliable scientific data and improved results. This cell was produced specifically so that any user can assemble the device without the requirement for prior experience or expertise. It is in the development of simple pieces like this that dramatically reduce the chance of user error. We feel that this is where AM can be beneficial to electrochemical research groups, not just those specialising in it, and therefore we next present some simple accessories to go alongside the cells and electrodes presented.

Additive manufacturing of laboratory accessories

One of the most important things in achieving reproducible and reliable electrochemical results is the electrochemical set-up. The ability to set up an experiment virtually identically every time with electrodes similarly spaced and key parameters such as the temperature recorded allows for added confidence in the reported results. AM can come into its own in this regard, allowing the researcher to quickly design, adapt and print small (or large) accessories to help with their own or fellow researchers experiments. In this section, we will highlight just a few such items we have produced and use regularly in our research group to improve the reliability of our results and help new students with their electrochemical set-ups. We reiterate that the STL files for these products are freely available for download from the Supporting Information Table 1 and we encourage others to come up with their own unique solutions to common laboratory problems, using recycled filament wherever possible.

As mentioned previously, a simple set-up with consistent electrode positioning is key when producing reliable electrochemical data. As such, we designed various clips that can help make this as simple as possible. This includes the lids for the electrochemical cells, as shown in Figs. 2A and 3A, which include holes for holding in place a thermometer, standard size commercially available reference and counter electrodes, commercial working electrodes and even one with a built in adapter for the use of SPEs. As can be seen in Fig. 3A, an additional measure for maintaining a tidy experimental set-up and reducing the chances of short-circuiting or damaging the wires connected to electrodes. The small wire holder slides onto a retort stand or clamp and can hold the three connections for the electrodes in place, removing the need for additional clamps, unnecessary bending of the wires or increased shielding from metal. In addition to this, we have produced a small piece that connects solidly onto the end of a standard retort stand clamp that can hold both the thermometer and commercial electrodes for use in a standard open electrochemical cell or beaker, Fig. 3B. This allows the user to maintain these experimental consistencies no matter what solvent, and therefore vessel, is being used.

Figures 3C and 3D show our simple and cost-effective design for SPE holders, designed for use in a clamp or for drop-casting on a benchtop respectively. These ergonomic designs, allow for simple set-ups depending on the experimental protocol being used and are extremely cost effective compared to commercially available SPE holders, costing £2.28 to produce including the cost of the edge connector and wire connections. In fact, we note that in some cases you could afford to buy a 3D printer and produce the SPE holder for less than some commercial SPE holders cost. The authors note that it is most likely impossible to produce one SPE holder that fits every commercial SPE, although this is where AM thrives as these designs can be easily adapted to include any edge connector needed, ensuring no researcher is limited to one specific brand. The SPE holders presented in this work utilise a low-cost edge connector (797-2132, RS Components) which fit standard SPE designs made in-house and used globally [61–63]. It is commonplace to drop-cast or perform droplet experiments using SPEs as the surface tension between aqueous solution and the SPE surface is typically strong enough to cover all three electrodes. When only wanting to functionalize the working electrode though, this can be more challenging and therefore AM can be used to create holders/moulds to make sure that only the desired electrodes are functionalized; Fig. 3E. An extended version of this for many more electrodes, which is sealed through the use of o-rings and magnets, can be found in the Supporting Information.

Finally, we include some general designs we have used that are not

Table 1

Summary of additively manufactured electrodes found in literature, how they were printed, their reported dimensions and intended applications.

ElectrodeMaterial	Printing Technique	Activation/Additives	Dimensions	Applications	Reference
Stainless steel	SLM	IrO_2	Helical; l: 1.5 cm; d: 0.5 cm	OER, pH sensing	[31]
TRGO	Binder Jet/Powder Bed	PdNP	Circular; d: 12 mm; t:300 μm	Supercapacitors	[64]
Stainless steel	Powder Bed	Ni coating	Square; 60 × 40 mm; t:3.7 mm	Electrochemical flow reactors	[65]
PLA/graphene (BM)	FFF	-	Circular; d: 17.75 mm; t: 1 mm Circular; d: 3 mm; t: 1 mm	Batteries, supercapacitors, HER	[33]
Stainless steel	SLM	Au plating	Helical; l: 1.5 cm; d: 0.5 cm	Phenol sensing	[66]
Stainless steel	SLM	Au plating	Helical; l: 1.5 cm; d: 0.5 cm	Nitroaromatic sensing	[67]
Stainless steel	SLM	Au plating	Helical; l: 1.5 cm; d: 0.5 cm	Heavy metal sensing	[68]
Stainless steel	SLM	Pt plating/ IrO_2	Gauze; not specified	HER, OER	[54]
PLA/graphene (BM)	FFF	DMF immersion	Ring; d: 15 mm; t: 3 mm Disc; d: 15 mm; t: 2 mm	Electroanalysis	[69]
Conductive PLA	FFF	Cu/graphite coating	Fractal	Solar Cell Counter Electrode	[34]
ABS		$NaMnO_2$		Batteries	[15]
PLA/graphene (BM)	FFF	$Ni\text{-}Fe_{10\%}$	Disc; d: 5 mm; t: 1 mm	OER	[35]
Stainless steel	SLM	TiO_2	'flag'; not specified	PEC water oxidation	[70]
PLA/graphene (BM)	FFF	DMF/acetone/EtOh/MeOH	Working area: 0.65 cm^2	Electrochemistry, supercapacitors	[41]
PLA/graphene (BM)	FFF	proteinase	Disc; d: 15 mm; t: 2 mm	1-naphthol sensing	[71]
PLA/graphene (BM)	FFF	-	Band; 0.08 × 0.15 cm	Flow cell, catechol sensing	[52]
PLA/carbon (PP)	FFF	GOx	Disc; ~5 mm	Glucose sensing	[72]
PLA/graphene (BM)	FFF	ZnO	Pyramid; 2.5 × 5 mm	batteries	[73]
PLA/carbon (PP)	FFF	NaOH activation	Disc; d: 0.54 cm; a: 0.23 cm^2	Dopamine sensing	[39]
PLA/graphene (BM)	FFF	Ag plating/bleach	Disc; d: ~5 mm	Reference electrode	[74]
PLA/NG	FFF	-	Disc; d: 3 mm; t: 1 mm	Heavy metal sensing	[27]
PLA/graphene (BM) PLA/carbon (PP) PLA/NG	FFF	EChem PBS activation NaOH activation -	Disc; d: 5 mm	NBOMes sensing	[26]
PLA/graphene (BM)	FFF	Thermal treatment (350°C)	Rectangle; a: 32.26 mm^2	Capacitors, batteries	[32]
PLA/nanocarbon	FFF	Thermal treatment (500°C)	Disc; d: 11 mm; t: 0.3 mm	Capacitors	[42]
PLA/Super P/2D-$MoSe_2$	FFF	-	Disc; d: 3.5 mm; t: 1.5 mm	HER/OER	[30]
PLA/graphene (BM)	FFF	DMF/EChem PBS activation	Disc; d: 8 mm; t: 1.6 mm	Mycotoxin sensing	[75]
PLA/graphene (BM)	FFF	DMF/EChem activation/AuNPs/HRP	a: 0.5 cm^2	H_2O_2 detection	[76]
PLA/NG	FFF	-	Disc; a: 0.22 cm^2	Manganese detection	[28]
PLA/graphene (BM)	FFF	MoO_2 nanowires	Disc; d: 6 mm; t: 1 mm	Electrochemical benchmarking	[77]
PLA/carbon (PP)	FFF	EChem Na_2SO_4 activation	Disc; d: 1.75 mm	Electrochemistry	[78]
PLA/graphene (BM)	FFF	EChem PBS activation	Disc; d: 0.18 cm^2	Cocaine sensing	[79]
PLA/LFP/CNT	FFF	-	Disc; d: ~1 cm; t: 0.3 mm	batteries	[29]
PLA/graphene (BM) PLA/carbon (PP)	FFF	EChem PBS activation/$NaBH_4$/ascorbic acid/ hydrazine/$LiAlH_4$ in THF	Disc; d: 11 mm; t: 0.3 mm	Electrochemistry	[45]
PLA/graphene (BM)	FFF	DMF/Echem	o-rin defined; d: 5.28 mm	Gunshot residue detection	[80]
PLA/graphene (BM)	FFF	EChem activation/PB	Disc; d: 5 mm; t: 1 mm	H_2O_2 detection	[81]
PLA/carbon (PP)	FFF	NaOH activation	Disc; d: 2.85 mm	Virus detection	[82]
PLA/carbon (PP)	FFF	NaOH activation	Disc; d: 5 mm	Drug screening	[83]

SLM: selective laser melting; l: length; OER: oxygen evolution reaction; TRGO: thermally reduced graphene oxide; PdNP: palladium nanoparticles; d: diameter; t: thickness; BM: Black Magic; HER: hydrogen evolution reaction; PP: Proto Pasta; NG: nanographite; a: area; AuNPs: gold nanoparticles; HRP: horseradish peroxidase; LFP: $LiFePO_4$; CNT: carbon nanotube; PB: prussian blue.

specifically for electrochemists, but are useful throughout the laboratory. Centrifuge tube holders, Fig. 3F, are extremely useful for work on biosensor development, electrode modification or aliquoting biological reagents to ensure the solutions are stored correctly at the bottom of the tubes and are easy to access. Similarly, Fig. 3G, shows a test tube holder which can also be used for larger centrifuge tubes. Both of these last designs can be easily modified depending on the size or amount of tubes needed, highlighting the flexibility and rapid prototyping ability of AM.

This work has highlighted just a few ways in which we use AM, not only for pushing the boundaries of AM forward, but also for improving our day to day experimental performance. We hope this has highlighted to the reader how AM can be beneficial to their laboratory environment.

Conclusions

In this manuscript, we have explored the area of AM and its use in the field of electrochemistry, (*viz.* Additive electrochemical manufacturing) highlighting how AM has been utilised in research, including the development of new conductive materials for printing, printing of electrodes, production of electrochemical cells and accessories to improve electrochemical experiments and teaching. AM and electrochemistry are an ever-expanding topic, with a majority of the work reported to date using commercially available filament for various applications. There is huge scope in the future for more bespoke printing materials to be made, with focus on other printing methods to FFF as well. For the development of electrochemical cells, there are numerous reports of how AM has been used to overcome significant challenges in

Table 2
Summary of additively manufactured electrochemical cells found in literature, how they were printed, what material they were printed from, their cell geometry and intended applications.

Electrochemical Cell Design	Printing Technique	Material Printed	Cell Geometry	Applications	Reference
Cylindrical Cell	FFF	PLA	Cylindrical – not specified	Water splitting	[54]
Electrochemical Flow Cell	FFF	PLA	Rectangular: 13.5 × 11.6 × 8.7 cm; 8 electrodes: 4.5 × 10.8 cm	NP synthesis	[57]
3 electrode cylindrical	FFF	ABS	Cylindrical; o-ring sealed WE (5.28/7.65 mm)	Electroanalysis	[51]
Electrolysis cell	FFF	ABS/translucent polymer	Rectangular base: ~7 cm length	Water splitting	[55]
Channel flow cell	FFF	PLA	Channel: 1.5 × 1 mm	Hydrodynamic electrochemistry	[52]
Fully AM Electroanalytical Cell	FFF	ABS	Cylindrical; volume: 5 mL	Electroanalysis	[39]
Wall-jet flow cell	SLA	Clear photopolymer resin (Clear FLGPCL02)	Cylindrical; volume 1.2 mL	HPLC-EC drug detection	[26]
3 electrode cell	FFF	Natural polyamide 12	3 necked, individual electrode chambers	Chemical reaction monitoring	[58]
2 chamber synthesis cell	Polyjet	Not specified	2 chambers; 6 × 5.25 mm for CE & RE; 4.5 × 1.5 mm for WE	Electrochemical NP conversion	[56]
3 electrode cell	FFF	ABS	Cuboid; 1.6 × 1.6 × 1 cm	Electroanalytical Hg detection	[49]
SPE cell	Polyjet	Polyacrylate resin	Cylindrical; 11 or 7 mm o-ring sealed	Impedimetric *E. coli* detection	[50]
Electrochemical Flow Cell	SLA	Form 2 Clear Resin (GPCLO4)	Rectangular body; 40 × 25 × 10 mm; Cell volume: 155 µL	HER scanning flow cell	[53]
Spectroelectrochemical/ Raman Cell	FFF	ABS	Cylindrical cell; d: 60 mm	Raman Spectroelectrochemistry	[59]
EChem/Raman Cell	FFF	ABS	Cylindrical cell; volume: 5 mL; PMMA transparent window 11.5 × 1.5 cm	Voltammetry/Raman of microparticles	[60]

FFF: fused deposition modelling; PLA: polylactic acid; NP: nanoparticle; ABS: acrylonitrile butadiene styrene; WE: working electrode; SLA: stereolithography; HPLC-EC: high performance liquid chromatography – electrochemistry; CE: counter electrode; RE: reference electrode; SPE: screen printed electrode; PMMA: poly(methyl methacrylate)

electrochemical set-ups. These have ranged from small cells designed for bulk solution analysis, to flow cells and fully AM water splitting cells. This highlights the advantage of having the ability to rapidly produce prototype cells in the lab, bespoke for a specific application. We have shown this can be taken further by highlighting how AM can be used to make accessories for electrochemical and other applications. These accessories, although basic, can greatly help with the electrochemical set-up, ensuring reliability and reproducibility in electrochemical data and also be extremely beneficial to new researchers to the field of electrochemistry. As such, we have provided all of the STL files for our designs in the Supporting Information Table 1 free to download. These files can be printed as is, preferably in recycled material, or modified using any CAD software to suit the researcher's specific needs. We hope other researchers find this useful and sparks their own ideas into how AM can be used beneficially throughout a laboratory environment.

Declaration of Competing Interests

None.

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Supplementary materials

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