

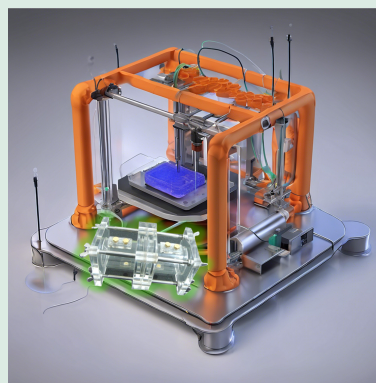
Leveraging 3D printing in microbial electrochemistry research: current progress and future opportunities

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HIGHLIGHTS

- 3D printing enables rapid prototyping and optimisation of MES reactors.
- 3D-printed electrodes improve electron transfer and biocompatibility.
- Tailored ink materials boost conductivity for sustainable energy.
- Bioprinting refines biofilm stability and microbial-electrode interactions.



ABSTRACT: Microbial electrochemical system (MES) offers sustainable solutions for environmental applications such as wastewater treatment, energy generation, and chemical synthesis by leveraging microbial metabolism and electrochemical processes. This review explores the transformative role of 3D printing in MES research, focusing on reactor body design, electrode fabrication, and bioprinting applications. Rapid prototyping facilitated by 3D printing expedites MES development while unlocking design flexibility, which enhances performance in optimising fluid dynamics and mass transfer efficiency. Tailored ink materials further improve the conductivity and biocompatibility of electrodes, paving the way for environmental applications. 3D-printed bio-anodes and bio-cathodes offer enhanced electrogenesis and boosted electron acceptance processes, respectively, by fine-tuning electrode architectures. Additionally, 3D bioprinting presents opportunities for scaffold fabrication and bioink formulation, enhancing biofilm stability and electron transfer efficiency. Despite current challenges, including material selection and cost, the integration of 3D printing in MES holds immense promise for advancing energy generation, wastewater treatment, resource recovery, carbon utilisation, and biosensing technologies.

KEYWORDS: 3D printing, Bioprinting, Microbial electrochemical system, Reactor body design, Novel electrode fabrication

1 Introduction

Microbial electrochemical system (MES) stands at the forefront of innovative solutions for wastewater treatment, resource recovery, and carbon utilisation, harnessing the power of microbial metabolism and electrochemical processes to fulfil environmental objectives (Aryal et al., 2022; Deng et al., 2023). By exploiting the unique ability of microorganisms to transfer electrons to solid surfaces, MES can efficiently degrade organic pollutants while simultaneously generating valuable products such as electricity, hydrogen, and value-added chemicals (Kong et al., 2023; Sharma et al., 2023; Harnisch et al., 2024). This dual functionality not only offers a sustainable approach to wastewater remediation but also holds promise for addressing energy demands and environmental pollution concerns (Fessler et al., 2024; Tang et al., 2024). In light of escalating global issues related to water scarcity and energy security, the development of MES technologies has garnered substantial attention as an environmentally friendly solution (Kundu et al., 2022; Yeung et al., 2024).

A typical dual-chamber MES reactor consists of an anode and a cathode chamber separated by a semi-permeable membrane, delineating two discrete compartments (Bhaduri and Behera, 2024). Within these compartments, electrochemically active bacteria drive the oxidation of organic matter at the anode and the reduction of a terminal electron acceptor (such as oxygen or carbon dioxide) at the cathode (Kumar and Kumar, 2022). What distinguishes MES from other bioreactors is its multifaceted structure, characterised by a multitude of components, each contributing crucially to its functionality. These components often incorporate electrodes, microbial biofilms, ion-selective membranes, and various supporting materials, rendering them inherently complex. This complexity stems from the necessity to optimise the properties and interactions of each component to achieve efficient electron transfer and maximise system performance (Chung et al., 2020; Jadhav et al., 2021). However, the traditional manufacturing methods used in MES construction, such as hand measuring, cutting, and bonding acrylic sheets, limit configuration options and result in coarse assembly, posing challenges for fine-tuning modifications to optimise performance (Jadhav et al., 2022; Garg et al., 2023).

3D printing technology, also known as additive manufacturing, has sparked a revolution across a multitude of industries by redefining traditional manufacturing processes (Johns, 2022; Parashar et al.,

2023). This innovative approach enables the fabrication of intricate three-dimensional objects layer by layer directly from digital designs (Williams and Sing, 2023). By employing extrusion, inkjet, or laser methods, 3D printing offers unparalleled freedom in creating complex geometries, internal structures, and customised components (Xu et al., 2021; Mousavi et al., 2023). The frequently-used 3D printing techniques based on these three methods are summarised in Table 1. As a result, 3D printing has permeated diverse sectors, including material science, engineering, medicine, biology, and environmental science (Kumar and Kumar, 2022). Its profound impact stems from its capacity to accelerate prototyping, diminish lead times, and enable on-demand production, thereby catalysing innovation and unlocking novel avenues for product design and development.

The advent of 3D printing has revolutionised the design and manufacturing of every constituent of MES reactors, offering unparalleled customisation and precision to address its multifaceted structure and intricate requirements: 1) it facilitates the creation of intricate reactor body designs with tailored features aimed at optimising fluid dynamics and mass transfer, thereby enhancing overall system performance (Chung and Dhar, 2021). 2) it enables the fabrication of electrodes with customised geometries and surface properties, thereby maximising electron transfer efficiency and current output (Xu et al., 2022b). 3) it allows for the precise fabrication of ion-selective membranes with desired properties, facilitating optimised ion transport while preventing the crossover of unwanted species (Jayathilake et al., 2022). 4) it empowers the fabrication of various supporting materials, including flow channels and structural elements, tailored to meet specific requirements and bolster the overall functionality and longevity of the MES system (Zhu et al., 2020; Wu et al., 2022). Through its versatility and adaptability, 3D printing emerges as a transformative tool in advancing the design and fabrication of MES components, heralding new possibilities for enhanced performance and functionality in MES (Chung and Dhar, 2021).

In this review, we explore the recent advances in the integration of 3D printing technology in MES manufacturing, focusing on its role in designing and optimising MES reactor bodies, electrodes, and scaffolds. We introduce how 3D printing enables rapid prototyping and accelerated development cycles, unlocking design flexibility and enhancing performance. Tailoring ink material properties allows for specific applications, while 3D-printed bio-anodes and bio-cathodes enhance

Table 1 Frequently used 3D printing techniques for reactor design and fabrication

3D printing type	3D printing technique	Abbr.	Principle
Extrusion-based 3D printing: These methods involve extruding material through a nozzle to build objects layer by layer.	Fused deposition modelling/ Fused filament fabrication	FDM/ FFF	Process: Melting and extruding thermoplastic filament. Materials: PLA, ABS, PETG, and other thermoplastics. Applications: Prototyping, functional parts.
	Direct ink writing	DIW	Process: Extruding a continuous filament of viscous material, such as paste or gel. Materials: Ceramics, hydrogels, conductive inks, biomaterials. Applications: Biomedical devices, electronics, custom manufacturing.
	Robocasting		Process: Extruding a ceramic slurry. Materials: Ceramics, pastes. Applications: Ceramics manufacturing, biomedical implants.
	Low-temperature deposition manufacturing	LDM	Process: Extruding materials at relatively low temperatures to build structures. This process is particularly suitable for temperature-sensitive materials. Materials: Hydrogels, biopolymers, certain plastics. Applications: Biomedical applications, electronics, temperature-sensitive prototypes.
Inkjet-based 3D printing: These methods use inkjet printer heads to deposit material in precise droplets.	Photopolymer Jetting	Polyjet	Process: Jetting layers of liquid photopolymer onto a build tray. The photopolymer droplets are immediately cured using UV light. Multiple layers are built up to create a solid object. Materials: Photopolymers, with options for different colours and material properties. Multiple materials can be combined in a single print. Applications: High-resolution prototypes, complex geometries, multi-material parts, dental and medical models, realistic prototypes with varying textures and colours.
	Binder Jetting	BJ	Process: Spraying a liquid binder onto a powder bed to fuse particles. Materials: Sand, gypsum, metal powders. Applications: Sand casting moulds, metal part prototypes, full-colour models.
	Material Jetting		Process: Jetting photopolymer droplets that are then cured layer by layer. Materials: Photopolymers, wax. Applications: High-resolution prototypes, dental and medical models.
	Drop-on-Demand	DOD	Process: Depositing droplets of build material in precise locations. Materials: Photopolymers, ceramics, metals. Applications: Precision manufacturing, electronics, microfabrication.
	Nanoparticle Jetting	NPJ	Process: Jetting liquid suspensions containing nanoparticles. The liquid evaporates leaving solid layers. Materials: Metals, ceramics. Applications: High-precision components for aerospace, medical, and industrial use.
Laser-based 3D printing: These methods use a laser to fuse or solidify material, usually in powder or liquid form.	Stereolithography	SLA	Process: Using a laser to cure liquid photopolymer resin layer by layer. Materials: Photopolymer resins. Applications: High-resolution prototypes, dental and medical models.
	Laser Powder Bed Fusion	LPBF	Process: Using a laser to selectively fuse powder particles on a bed, building up layers to form a solid part. Materials: Metals such as aluminium, titanium, and steel. Applications: Aerospace, automotive, medical implants, complex metal parts.
	Selective Laser Sintering	SLS	Process: Using a laser to sinter powder particles together. Materials: Nylon, polyamide, other polymers. Applications: Functional prototypes, end-use parts.
	Selective Laser Melting	SLM	Process: Using a laser to fully melt metal powder particles. Materials: Aluminium, titanium, steel. Applications: Aerospace, medical implants, complex metal parts.
	Laser-Induced Forward Transfer	LIFT	Process: Using a laser to transfer material from a donor substrate to a receiver substrate. The laser pulse causes a small amount of material to be ejected from the donor and deposited onto the receiver. Materials: Metals, biomaterials, polymers, and other specialised inks. Applications: Electronics, sensors, biomedical applications, and microfabrication.
	Digital Light Processing	DLP	Process: Using a digital light projector to cure liquid photopolymer resin. Materials: Photopolymer resins. Applications: High-resolution and fast prototypes, dental models.
	Continuous Liquid Interface Production	CLIP	Process: Using a continuous UV light source to cure liquid resin while pulling the object from the resin bath. Materials: Photopolymer resins. Applications: High-speed, high-resolution parts.

electrogenesis and electron acceptance processes. Additionally, we discuss the applications of 3D bioprinting in MES, including scaffold fabrication and bioink formulation. Lastly, we address current

challenges and opportunities, emphasizing the transformative potential of 3D printing in advancing MES technology for environmental remediation and energy generation.

2 3D printing-assisted design and optimisation of MES reactor

The utilisation of 3D printing technology holds significant promise for the design and optimisation of MES reactors, primarily due to its transformative impact on accelerating innovation and addressing the inherent complexities of MES architectures (Dwivedi et al., 2022; Verma et al., 2024). The adoption of 3D printing expedites the design-build-test cycle, allowing for rapid prototyping and iterative refinement of reactor configurations (Trauger et al., 2022). This iterative process enables researchers to explore a wide range of geometries and configurations, ultimately optimising reactor performance with enhanced efficiency and precision. Moreover, 3D printing facilitates the fabrication of complex reactor structures that surpass the limitations of conventional manufacturing techniques (Zhakeyev et al., 2021). By leveraging 3D printing, one can create intricate and customised designs tailored to the specific requirements of MES applications, thereby enhancing functionality and performance (Jayathilake et al., 2022). Furthermore, the sustainable nature of 3D printing ink materials contributes to eco-conscious manufacturing practices, aligning with the growing emphasis on sustainability within MES research and development (Chen et al., 2022b; Abbas et al. 2024). In essence, the integration of 3D printing technology represents a pivotal advancement in the field of MES, offering unparalleled opportunities for innovation and optimisation to meet the evolving demands of bioelectrochemical processes.

2.1 Unlocking design flexibility and enhanced performance

3D printing enables the production of complex fluidic structures unattainable by other methods, while printed reactors offer scalability, facilitating size adjustments based on specific reaction parameters (Griffin and Pappas, 2023; Duarte et al., 2024). For example, Poly-Jet 3D printing can mass-print and directly UV-cure highly porous monolithic enzyme carriers with various specifications for immobilising enzymes, enabling the fast and simple implementation of enzyme cascades and effective enzyme transformation modules (Siripongpreda et al., 2023; Zhang et al., 2023). Another representative example lies in the filter-press electrochemical reactor (Fig. 1(a)), where 3D printing was used to obtain critical components of the device, including flow frames, detachable manifolds, turbulence promoters, and current collector holders (Márquez-Montes et al.,

2020). The 3D-printed materials demonstrated satisfactory chemical stability and compatibility, and the designed configurations and geometries showed enhanced turbulence and mass transfer coefficients, suitable for processes like electrosynthesis, electrochemical water splitting, and bioelectrochemical processes.

While research has extensively explored innovating 3D-printing-based reactor designs to optimise system performance in enzyme engineering, electrochemical, or photocatalysis processes (Aguirre-Cortés et al., 2023; Remonatto et al., 2023; Tan et al., 2023), practical implementation in MES remains limited. This can be attributed to the inherently complex nature of MES, which involves various interacting components together with the microbes, making it challenging to implement comprehensive performance enhancement (Arun et al., 2024; Cai et al., 2024). Currently, prevalent applications mainly centre around biosensors and MFCs, where reactor configurations tend to be relatively simpler. For instance, recent studies have demonstrated the fabrication of sensor structures using compositing materials of conductive polymers, metals, and carbonaceous materials enabled by 3D printing, broadening the scope and affordability of electrochemical biosensors in healthcare diagnostics (Elbadawi et al., 2021). Another study demonstrates a fully 3D-printed impedance-based biosensor with reduced testing time for antibiotic susceptibility, offering a cost-efficient diagnostic tool for rapid assessment (Domingo-Roca et al., 2023). Besides, the flow-through cell (Fig. 1(b)) is a typical example of applying 3D-printed biosensors for real sewage systems through electrochemical determination of harmful pharmaceuticals (Dakošová et al., 2023). These examples highlight the tangible benefits of integrating 3D printing into biosensor development, emphasizing its potential impact within MES applications. Additionally, 3D-printed flow cell systems can also characterise biofilm growth using electrochemical impedance spectroscopy (EIS) (McGlennen et al., 2023). By employing microfabricated sensors featuring gold micro-interdigitated electrodes modified with an electrically conductive polymer layer, the study achieved highly stable, time-resolved EIS measurements with reduced variability compared to unmodified sensors. Through parallel characterisation with high-resolution confocal laser scanning microscopy, distinct changes in EIS data corresponding with consistent biofilm growth stages were observed, highlighting the potential of 3D-printed flow cell systems in studying dynamic biofilm processes within MES environments.

Expanding on the applications of 3D printing in MFC technology, recent research has showcased its

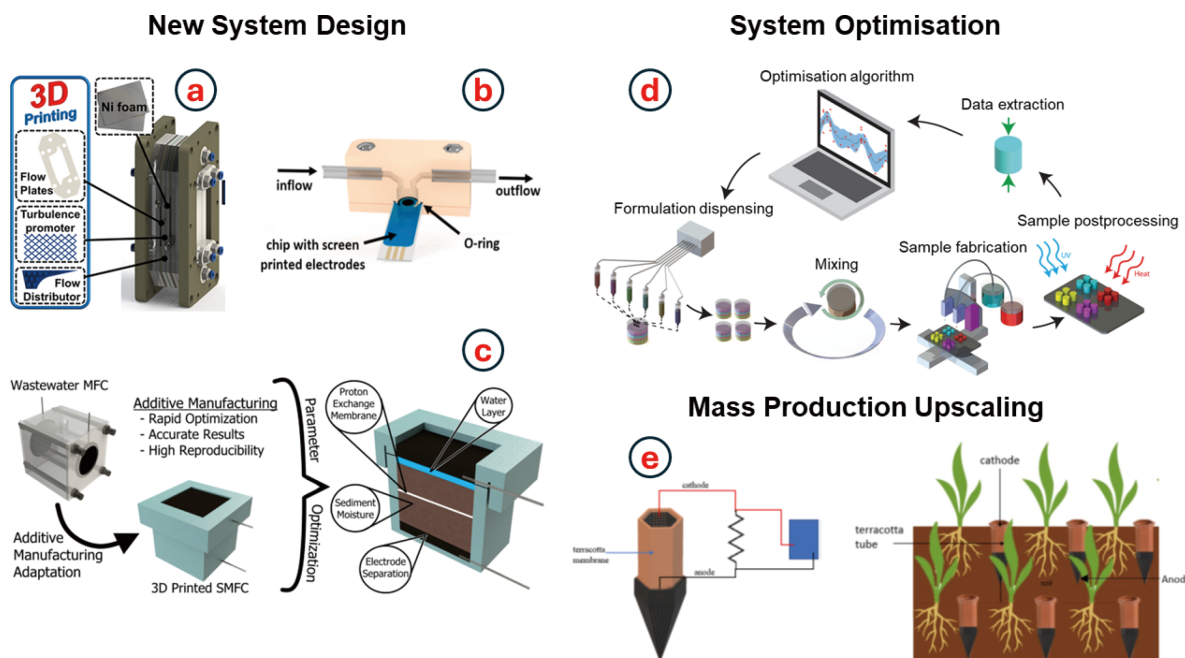


Fig. 1 3D printing application to MES reactor design in terms of 1) new configurations (a) filter-press reactor, copyright 2020, American Chemical Society (Márquez-Montes et al., 2020); (b) flow-through cell, copyright 2023, Elsevier (Dakošová et al., 2023); (c) MFC, copyright 2023, Springer Nature (Kim and Simson, 2023); 2) (d) system optimisation, copyright 2021, licence 4.0 (CC BY-NC) (Erps et al., 2021); and 3) (e) mass production upscaling, copyright 2023, licence 4.0 (CC BY-SA) (Constantino et al., 2023).

versatility in optimising MFC performance through innovative designs and customised components (Dwivedi et al., 2022; Kamali et al., 2023). For instance, 3D printing has enabled the fabrication of intricate electrode structures with enhanced surface area, leading to improved electron transfer kinetics and overall MFC efficiency (Sreelekshmy et al., 2020). By precisely controlling the geometry and porosity of the electrodes, especially the combining printing of membranes in porous filaments and electrodes made of conductive PLA filaments, researchers have achieved significant enhancements in power output and current density (You et al., 2017). These 3D-printed MFCs with the membrane electrode assembly were recently utilised for urine wastewater treatment and successfully scaled up through the stacking of modules (Chang et al., 2023). Moreover, 3D printing has facilitated the development of specialised reactor bodies and membranes tailored to MFC operation, allowing for better control over key parameters such as flow dynamics and microbial colonisation. For instance, in a recent study, a new 3D-printed sediment MFC was designed (Fig. 1(c)) to investigate the effects of sediment moisture, water overlayer, proton exchange membrane usage, and electrode distance on system power output (Kim and Simson, 2023). After iterative

modifications, the system was successfully employed for electricity generation from practical urban stream sediment, demonstrating the versatility and effectiveness of 3D printing in optimising MFC performance for real-world applications. Additionally, the scalability of 3D printing has enabled the mass production of MFC units with consistent quality, paving the way for large-scale implementation in various settings (Yeo et al., 2024). By leveraging the design flexibility and precision offered by 3D printing, researchers continue to push the boundaries of MFC technology, unlocking new opportunities for sustainable energy generation and environmental remediation.

For complex MES systems, however, current research primarily focuses on optimising specific components rather than pursuing overall configuration innovation. For example, in Corral's study, the optimised 3D-printed sparging chamber connecting cathode gas and catholyte compartments enabled the effective saturation of CO_2 in the electrolyte, minimising pH gradients and concentration polarisation, and preventing breakthroughs of electrolyte and flooding (Corral et al., 2021). Through iterative improvements informed by metrics such as total cell resistance, successive generations of reactors demonstrated enhancements in improved faradaic

efficiencies, and achieved high current densities exceeding 500 mA/cm², highlighting the potential of 3D printing for high-rate electrosynthesis from CO₂.

2.2 Rapid prototyping and accelerated development cycles

The development cycle of MES reactors is significantly accelerated by 3D printing through rapid prototyping and iterative design exploration (Rajesh and Kumawat, 2024). This technology empowers researchers to leverage computer-aided design software for the swift creation and modification of reactor designs, which facilitates the optimisation of fluid dynamics and mass transfer within the reactor (Khoo et al., 2024; Montaner and Hilton, 2024). Unlike traditional manufacturing methods, 3D printing enables the creation of intricate geometries and tailored features with remarkable speed and precision. This translates to faster testing cycles for new designs and configurations, allowing researchers to evaluate performance and make iterative improvements (Fig. 1(d)) (Erps et al., 2021). Additionally, 3D printing offers substantial cost and time savings compared to conventional techniques (Praveena et al., 2022). This reduction in resource expenditure allows for the development of multiple design iterations for experimentation. A compelling example is a study by Corral et al., where stereolithography (SLA) 3D printing was employed to create multiple iterations of laboratory-scale, vapor-fed electrochemical systems for CO₂ reduction (Corral et al., 2021). This approach resulted in a 100-fold decrease in production costs and time for reactor fabrication compared to traditional manufacturing methods, highlighting the pivotal role of 3D printing in advancing MES technology toward commercialisation.

In terms of MES upscaling, the impact of 3D printing encompasses critical stages such as reactor manufacturing, bioelectrochemical performance testing, and scalable reactor design engineering. For example, 3D printing can significantly enhance the development of bioelectro-methanation processes by expediting rapid prototyping, optimising performance, and facilitating comprehensive techno-economic evaluations (Jayathilake et al., 2022). Notably, the versatility of 3D printing enables the development of scaled devices for diverse biogas sources, including those found in dairy farms, crop residue processing, food waste anaerobic digestion facilities, and wastewater treatment plants (Baena-Moreno et al., 2021; Cioabla et al., 2022). Another relevant application lies in the field of microbial fuel cells (MFCs), where large-scale implementation faces challenges due to the dispersed

nature of power generation (Mukherjee et al., 2022, Neethu et al., 2022). Traditionally, there has been no clear path to consolidate or amplify the small power output of individual MFCs. To address this hurdle, a recent study investigated stacking configurations of 3D-printed MFCs (Fig. 1(e)) to determine their potential for amplifying bioelectricity generation (Constantino et al., 2023). In their setup, the electrodes were 3D printed using polylactic acid (PLA) filament and terracotta membranes were used as separators. Six cells were successfully built with electrodes designed for a snug fit with the ceramic separators. This research exemplifies the promising role of 3D printing technology in upscaling MES applications and offers valuable insights into optimising stacking configurations for enhanced power generation efficiency.

2.3 Tailoring ink material properties to specific applications

In the realm of 3D-printed MES, the selection of printing ink and techniques plays a crucial role in shaping the performance and durability of reactors. 3D printing ink materials encompass a diverse array of substances, each tailored to meet specific application requirements (Guzzi and Tibbitt, 2020; Saadi et al., 2022). Common materials include thermoplastics like PLA, acrylonitrile butadiene styrene (ABS), and polyethylene terephthalate glycol (PETG), prized for their ease of printing, durability, and versatility (Gregor-Svetec, 2022). These thermoplastics are often utilised in methods like fused deposition modelling (FDM) or fused filament fabrication (FFF) due to their cost-effectiveness and chemical stability (Daminabo et al., 2020). However, challenges such as potential leakage may arise due to the extrusion-like application of layers, particularly if not well-bound to surrounding layers (Kim and Nam, 2020, Tian et al., 2021). To address this, SLA presents an alternative where previous layers are chemically bonded, allowing for higher geometrical resolution (Li et al., 2022a). Yet, UV-resin materials used in SLA may exhibit brittleness and degradation over time, posing durability concerns in MES applications (Gao et al., 2023). In this case, for thermally demanding MES reactions, selective laser melting (SLM) of metals emerges as a promising solution. By utilizing stainless steel via SLM, reactors can be fabricated with exceptional thermal conductivity and mechanical strength (Hauser et al., 2022; Sharma et al., 2024). This enhances their ability to withstand the rigours of MES processes. In addition to thermoplastics and metals, thermosetting polymers such as epoxy resins and polyurethanes offer enhanced mechanical strength and chemical resistance

post-curing, making them suitable for specific MES applications requiring robustness and durability (Yuan et al., 2020; Liu et al., 2023; Xie et al., 2024a).

For applications in MES, where the interaction between microorganisms and electrodes is pivotal, ink formulations can be tailored to optimise conductivity, biocompatibility, and chemical stability (Bastola et al., 2023; Kumi et al., 2024). Metal-based inks, comprising metal powders dispersed in a binder, enable the fabrication of metallic parts with varying degrees of conductivity and strength (Ramazani and Kami, 2022). Meanwhile, ceramic inks composed of ceramic particles suspended in a liquid medium facilitate the printing of heat-resistant, electrically insulating components (Abdolmaleki et al., 2021; Li and Huang, 2024). Conductive additives like carbon nanotubes or graphene enhance electrical conductivity, promoting efficient electron transfer between microbes and electrodes (Yi et al., 2020; Wang et al., 2021). Biocompatible polymers such as hydrogels or bioresorbable materials ensure compatibility with microbial growth and proliferation, minimising adverse effects on microbial activity (Kirillova et al., 2021; Mallakpour et al., 2021). Chemical modifications, such as surface functionalisation or doping, can further fine-tune material properties to suit MES requirements, facilitating tailored interactions between microbes and electrodes for enhanced performance and stability (Yaqoob et al., 2020; McCuskey et al., 2022; Krasley et al., 2024). Overall, the versatile nature of 3D printing ink materials enables the precise customisation of material properties, enabling the development of innovative solutions for microbial electrochemical reactors and beyond.

3 3D printing for conductive and biocompatible electrode design and fabrication

The impetus for integrating 3D printing with microbial electrochemistry arises from the compelling synergy between these pioneering technologies, poised to address challenges in wastewater treatment and resource recovery (Saidulu et al., 2022; Kamali et al., 2023). While MESs offer a sustainable approach to wastewater remediation and energy generation, they encounter obstacles such as inefficient electrode designs and biofilm formation (Greenman et al., 2021; Varjani, 2022). Leveraging 3D printing capabilities, researchers can meticulously tailor electrode architectures, exerting precise control over surface properties, porosity, and topology to optimise microbial adhesion

and electron transfer kinetics (Al-Amiery et al., 2024; Guo et al., 2024). 3D printing allows for the deposition of functional coatings and materials onto electrode surfaces, enhancing their electrochemical performance and biocompatibility (Browne et al., 2020; Jiang et al., 2020). This includes the incorporation of conductive polymers, nanomaterials, and bioactive compounds to promote electron transfer and microbial adhesion (Maziz et al., 2021; Peñas-Núñez et al., 2023). Additionally, electrode design and fabrication benefit from 3D printing, as it enables precise control over architecture and geometry, facilitating the creation of complex structures tailored to enhance microbial adhesion and electron transfer (Chen et al., 2023). This includes the integration of hierarchical surface features and microfluidic channels to optimise mass transport and biofilm formation (Li et al., 2022b; Roy Barman et al., 2023).

In MES, the anode and cathode play distinct yet interconnected roles, driving various processes for different purposes. The anode serves as the site of microbial oxidation, where microorganisms oxidise organic compounds or other substrates, releasing electrons in the process (Prathiba et al., 2022; Klein et al., 2023). These electrons are then transferred through an external circuit to the cathode. Simultaneously, protons are released, migrating through a conductive medium to the cathode. At the cathode, these electrons and protons combine with an electron acceptor, typically oxygen or another compound, leading to reduction reactions. This reduction facilitates the overall electron flow in the system, generating an electrical current (Mohyudin et al., 2022). MES finds applications in wastewater treatment, bioenergy production, biosensor development, and the synthesis of valuable chemicals, showcasing the versatile roles of the anode and cathode in harnessing microbial activity for various purposes (Wang et al., 2020; Chu et al., 2021; Yi et al., 2022; Gupta et al., 2024). Figure 2 summarises the construction of different bioanodes and biocathodes in MES using different 3D printing techniques.

3.1 3D-printed anode for enhanced electrogenesis processes

Bioanodes are the key component of MES for harnessing the power of microorganisms to convert organic matter into electricity or other valuable products (Yaqoob et al., 2021). The advent of 3D printing has revolutionised bioanode design, facilitating the creation of intricate structures with bespoke properties crucial for enhancing electron transfer from bacteria to the electrode surface (Pankan et al., 2020; He et al., 2021b).

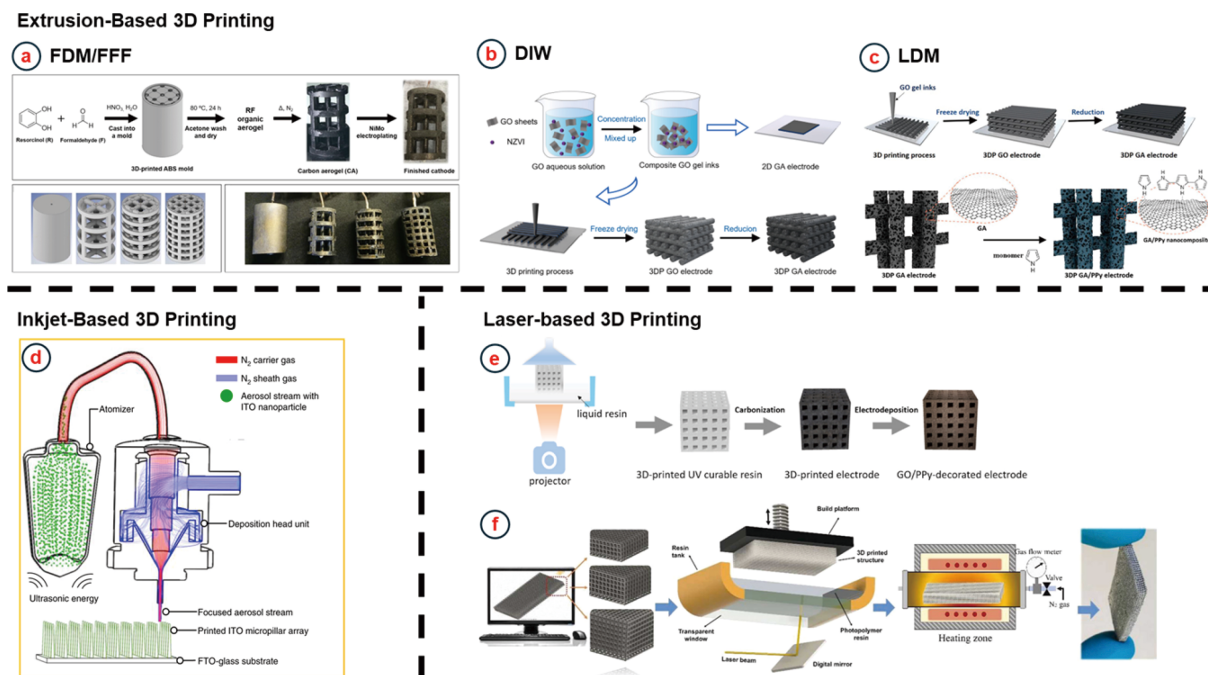


Fig. 2 Different 3D printing techniques applied in electrodes manufacturing in MES: (a) FFF-printed NiMo-plated carbon aerogel cathode, copyright 2021, licence 4.0 (CC BY) (Kracke et al., 2021); (b) DIW-printed NZVI-functionalised GA bioelectrode, copyright 2024, Elsevier (He et al., 2024); (c) LDM-printed hierarchical porous GA/PPy biocathode, copyright 2023, Elsevier (He et al., 2023); (d) Aerosol-jet-printed FTO-coated micropillar photoelectrode, copyright 2022, Springer Nature (Chen et al., 2022a); (e) SLA-printed hierarchical porous GO-Ppy bioanode, copyright 2021, Elsevier (He et al., 2021a); (f) SLA-printed reticular pyrolytic carbon cathode, copyright 2024, Elsevier (Zou et al., 2024).

Across various studies, a spectrum of 3D printing techniques has been deployed to fabricate bioanodes, tailored to diverse conductive electrode materials for different microbial electrochemical processes, such as electricity and hydrogen production, hydrogenotrophic denitrification, and bio-photoelectrochemical processes. Representative techniques, as outlined in Table 2, encompass extrusion-based printing for carbon-based, metal printing-based, as well as 3D-printed anodes imbued with biocompatibility and photoelectroactive properties.

Extrusion-based printing has demonstrated effectiveness in fabricating carbon-based bioanodes, including graphene oxide/polypyrrole (GO/PPy) scaffold, graphene aerogel (GA), and PLA/graphene bioanodes (He et al., 2021a; 2021b; Alonso et al., 2022). The precise deposition and shaping capabilities of extrusion-based printing enable the creation of complex structures with high surface areas and hierarchical pore architectures, facilitating efficient nutrient supply and waste dissipation within and outside the anodes. Notably, periodic macropores within these structures provide open channels for optimising nutrient flow and waste removal, while micropores derived from the graphene network significantly enhance the surface area

available for bacterial colonisation and proliferation. These structural features are paramount for maximising bacterial attachment and substrate diffusion, thereby enhancing bioelectrocatalytic performance.

Fabricating metal-based bioanodes poses unique challenges due to their inherent stiffness and processing difficulties, which 3D printing techniques have emerged as promising tools to address. Laser-based powder bed fusion (L-PBF) is a unique approach for fabricating metal bioelectrodes, utilizing a laser to selectively melt and fuse together layers of metal powder, enabling the creation of complex 3D metal structures with high precision. For instance, in the fabrication of an octahedral lattice-structured stainless-steel anode, precise control over geometry offered by L-PBF method enabled the creation of an optimised lattice design with a 3.0 mm pitch, resulting in a significant improvement in current production (29.5 A/m^2) compared to conventional carbon-felt electrodes (Yamashita et al., 2023). Another study focused on copper-based bioanodes, utilizing FDM printing of copper-based Electrifi conductive filaments to fabricate electrodes with various spiral geometries. The 1-cycled spiral design exhibited superior performance, with a 2.6-fold higher current density and a 5-fold faster

Table 2 Representative innovative 3D-printed anodic electrodes applied in MES for multiple purposes in recent three years

3D-printed anode	Process	Manufacturing method	Aim	Results	Reference
Hierarchical porous GO/PPy bioanode	Power production	SLA photosensitive resin ink, layer-by-layer printing (iSLA1900D), carbonisation	Enhance the biofilm formation and electron transfer at the biologic/inorganic interface	Highest power density of $22.4 \pm 0.6 \text{ W/m}^2$	He et al. (2021a)
GA bioanode		GO gel ink, LDM printing (BP11), ice crystallisation and freeze-dried	Increase the bacterial colonisation and proliferation surface area of each printed wire	Highest volumetric current density of $10608 \pm 1036 \text{ A/m}^3$	He et al. (2021b)
PLA/graphene bioanode		Fused polymeric compound, graphene-loaded PLA with FDM printing (Prusa i3), electrochemical activation	Suitability of 3D-printed bioanodes and the effects of activation methods	Maximum current density reached 5 A/m^2	Alonso et al. (2022)
Octahedral lattice-structured stainless-steel anode		L-PBF method with the metal 3D printer (OPM250L), Spherical powders were laminated using a Ytterbium fibre laser, heat treatment	Form an iron-oxide layer on the electrode surface for increasing biocompatibility, and optimise pitch length to improve surface area and substrate diffusion	Maximum current density of 29.5 A/m^2 obtained with the 3.0 mm-pitch electrode, volumetric current of 1.18 mA/cm^3 bounding box	Yamashita et al. (2023)
Spiral metal anode	Hydrogen production	Copper-based Electrifi filament (Multi3D)	Better contact between the electrode-electrolyte interface and greater vertical flow control to facilitate electron exchange	Maximum current density of 1.5 mA/cm^2 at 2 V, 2-fold faster than control electrodes	Baş and Kaya (2022)
Corncob-filled sandwich structure anode	Bioelectrochemical power denitrification	Composed of corncob, porous carbon felt anodes, and 3D-printed metal current collector	Sandwich structure anode to control carbon source release and enhance electron collection	Higher denitrification efficiency ($21.79 \pm 0.22 \text{ g NO}_3^- \text{-N}/(\text{m}^3 \cdot \text{d})$) than control system	Li et al. (2023)
FTO-coated micropillar photoelectrode	Biophoto-electrochemical power production	ITO precursor with water and methanol, micropillar structures fabricated with aerosol jet printer (AJ200)	Generate micropillar array structures with multiscale hierarchical features using nanoparticles, and increase roughness to boost electrode surface area, cell loadings and light collection	The photocurrent outputs up to $245 \mu\text{A/cm}^2$ for mediated electron transfer, and up to $1.93 \mu\text{A/cm}^2$ for direct electron transfer	Chen et al. (2022a)

hydrogen production rate compared to other geometries (Baş and Kaya, 2022). These results underscored the critical role of electrode geometry in optimising electron transfer and fluid dynamics, showcasing the potential of 3D printing for fabricating complex metal-based bioanodes with tailored properties.

3D printing techniques have also demonstrated their capability to integrate biocompatible or photo-electroactive materials into conductive bioanodes. For the development of a corncob-filled sandwich structure electrode, agricultural waste corncob served as a cost-effective carbon source for electron generation within the sandwich structure anode (Li et al., 2023). The construction and optimisation of 3D-printed anode structure were guided by COMSOL software to regulate carbon source release and enhance electron collection, achieving a significantly higher denitrification efficiency ($21.79 \pm 0.22 \text{ g NO}_3^- \text{-N}/(\text{m}^3 \cdot \text{d})$) compared to control systems. In another study, aerosol jet printing was employed to fabricate micropillar array electrodes incorporating indium tin oxide nanoparticles (Chen et al., 2022a). This innovative method allowed for the creation of tuneable hierarchical features across five orders of magnitude in length scales in one printing step, facilitating the production of libraries of hierarchical micropillar array electrodes. The resulting hierarchical pillar structure, featuring micro branches,

contributed to increased submicrometric roughness on the surfaces, thereby enhancing the electroactive surface area, cell loadings, and light collection efficiency. The photocurrent density was consequently elevated to $245 \mu\text{A/cm}^2$, nearly doubling the performance achieved by previous methodologies. This study underscores the immense potential of 3D printing in fabricating intricate bio-electrode interfaces tailored for semi-artificial photosynthesis, thus paving the way for enhanced solar energy conversion.

3.2 3D-printed cathode for boosted electron acceptance processes

In contrast to bioanodes that harvest electrons from organics catalysed by electroactive microorganisms, biocathodes serve as the terminal of the electron flow loop in MES, providing the necessary surface and conditions for oxidising agents to accept electrons, including carbon dioxide, nitrate, and oxygen, and thereby facilitating the bioelectrochemical processes including advanced oxidation, methane or volatile fatty acids production, autotrophic denitrification, and electricity generation (Noori et al., 2020; Zhao et al., 2021; Su and Chen, 2022; Mo et al., 2023). Across various MES applications, 3D printing offers a versatile toolbox for crafting high-performance biocathodes. Studies

have explored a range of 3D-printable materials and techniques, each targeting specific functionalities to enhance MES processes (summarised in Table 3). This tailored design approach stands in stark contrast to traditional biocathode fabrication methods, which often lack precise control over crucial properties.

The traditional 3D printing techniques employed for biocathode fabrication include extrusion-based, inkjet-based, and laser-based SLA printing. As described in Table 1, extrusion-based 3D printing involves depositing a continuous filament of material through a nozzle, creating intricate 3D structures layer by layer (Daminabo et al., 2020). This technique was recently used for fabricating biocathodes utilizing a variety of materials, including conductive polymers and carbon-based materials. Conductive polymers, such as PLA, polyaniline, and ABS, offer biocompatibility, tuneable conductivity, and the ability to incorporate functional additives (Menzel and Tudela, 2022). For example, a nickel-based coated conductive PLA lattice biocathode was designed explicitly for stable microbial electrosynthesis of acetate and methane from CO₂ (Nwanebu et al., 2022). The electrode was 3D printed to enhance the surface area for microbial attachment, followed by metal electrodeposition (Ni, Fe, Mn) to improve electromethanogenesis performance. A significant enhancement in hydrogen evolution kinetics on the cathode surface resulting from the more efficient

electron transfer was recognised to drive the desired biocatalytic reactions for methane and acetate production at close to 100% coulombic efficiency, which presented a more than 3-fold increase in CH₄ and acetate production rates compared to the bare PLA control cathode. In line with the conductive polymers, carbon-based materials, such as graphene aerogel and carbon felt, were also employed to fabricate biocathodes due to their excellent electrical conductivity and high surface area for microbial attachment. For example, the NiMo-plated carbon aerogel cathode was fabricated using the FFF printing approach to enhance electromethanogenesis performance in an MES, where the resorcinol-formaldehyde gels served as the filament precursor, and the custom ABS mould was used to shape the extruded filament during the printing process (Kracke et al., 2021). The 3D-printed design facilitated a significant increase in surface area for improved microbial attachment, resulting in a 2.14-fold boost in methane production compared to a traditional carbon felt cathode, highlighting the potential of 3D printing with tailored materials for optimising MES applications. Alongside the thermal FFF method, low-temperature liquid deposition modelling (LDM) also offers promise for fabricating carbon-based cathodes in electromethanogenesis. He et al. (2023) demonstrated this with their hierarchical porous GA/PPy biocathode, achieving a high CH₄ production rate (1672 ± 131

Table 3 Representative innovative 3D-printed cathodic electrodes applied in MES for multiple purposes in recent three years

3D-printed cathode	Process	Manufacturing method	Objective	Results	Reference
Reticular pyrolytic carbon cathode	Advanced oxidation	SLA (Preform), high-temperature resin precursors followed by pyrolysis	Increase the electroactive surface area of and boost the 2e ⁻ ORR kinetics	H ₂ O ₂ production of 129.2 mg/L in 12 h, 2.3–6.9 times greater than conventional electrodes	Zou et al. (2024)
NiMo-plated carbon aerogel cathode	Electromethanogenesis	Resorcinol and formaldehyde gels base, custom ABS mould, FFF printer (Lulzbot Taz 6), carbonised at high temperature	Mitigate effects of bubble formation and local pH gradients within the boundary layer and facilitate <i>in situ</i> electron delivery	CH ₄ production rate of 2.2 L/(Lcatholyte · d) and at a coulombic efficiency of 99%	Kracke et al. (2021)
Hierarchical porous GA/PPy biocathode		GO gel ink, low-temperature LDM printing (BP11), ice crystallisation and freeze-dried	Facilitate both interior biofilm formation and mass transfer, and provide a large surface area for microbial colonisation	CH ₄ production rate of 1672 ± 131 mmol/(m ³ · d)	He et al. (2023)
NZVI-functionalised GA bioelectrode	Solar-driven electromethanogenesis	DIW printing (BP11), Graphene inks containing 1.79 mmol/L NZVI, ice crystallisation, freeze-drying	Enhanced high specific surface area for microbial colonisation while ensuring mass transfer. NZVI can assist <i>in-situ</i> hydrogen generation, facilitating biofilm formation and CH ₄ production in the indirect electron transfer pathway.	Maximum CH ₄ production rate of 6993 ± 832 mmol/(m ² · d) with a faradaic efficiency of 83.7%, 3.24-fold to the carbon felt electrode, and solar-to-CH ₄ efficiency of 4.70%.	He et al. (2024)
Nickel-based coated Conductive PLA lattice cathode	Bioelectrosynthesis to acetate and CH ₄	Conductive PLA filament to form a lattice for electrode printing (Prusa I3 MK2), sonication, deposition with NiFeMn catalyst	Metal-coated cPLA lowers electrical resistance, and enhances electron transfer between the cathode and the biofilm	At 2.8 V, maximum production rate of CH ₄ 50 ± 6 mL/d, acetate 185 ± 27 mg/d, and H ₂ 545 ± 175 mL/d at close to 100% Coulombic efficiency	Nwanebu et al. (2022)
Alginate-based activated carbon cathode	Power production	Preloaded with conductive paste, extrusion using syringes on the EvoBot	Discover eco-friendly and printable air cathode	Power production of 286 μW	Theodosiou et al. (2020)

mmol/(m² · d)). The interconnected channels within the GA structure facilitated efficient mass transfer of nutrients, gases, and electrons throughout the biofilm, while the PPy coating improved biocompatibility and enhanced electron transfer between the microbes and the electrode.

Inkjet-based 3D printing utilises a piezoelectric or thermal inkjet nozzle to deposit droplets of material, enabling precise patterning and high-resolution structures (Bastola et al., 2023). This technique is particularly suitable for fabricating biocathodes with complex geometries and incorporating multiple materials (Blyweert et al., 2021; Zub et al., 2022). For example, a 3D-printed GA biocathode was constructed by inkjet printing and afterwards functionalised with nano zero-valent iron (NZVI) to catalyse solar-driven electromethanogenesis process. The GA material featured hierarchical pores that significantly increased the specific surface area for microbial growth, while the NZVI played a crucial role in producing hydrogen *in situ*. Together, both of these aided in forming functional biofilms and promoting CH₄ production through an indirect electron transfer pathway (He et al., 2024). A maximum CH₄ production rate of ~7 mol/(m² · d) with a faradaic efficiency of 83.7% was achieved, which was 3.24 times higher than that of a traditional carbon felt electrode. Besides, when the biocathode was incorporated into a PV-electrolyser cell, it achieved a solar-to-CH₄ efficiency of 4.70%. SLA-based 3D printing involves curing a photosensitive resin layer by layer using a laser or light source, providing high-resolution and smooth-surfaced biocathodes ideal for applications requiring precise control over electrode morphology (Maines et al., 2021; Ashammakhi et al., 2022). Recently, SLA was employed to fabricate a 3D pyrolytic carbon (PyrC) cathode for advanced oxidation, achieving a maximum H₂O₂ production of 129.2 mg/L in 12 h with a faradaic efficiency of 94%, surpassing conventional electrodes by 2.3–6.9 times (Zou et al., 2024). Furthermore, the proposed 3D PyrC electrode exhibited exemplary scalability, reusability, and mechanical properties, affirming its practical suitability for large-scale applications. Beyond H₂O₂ synthesis, its application in the bio-electro-Fenton process highlighted its effectiveness as a tertiary treatment method for eliminating micropollutants.

4 Applications of 3D bioprinting in MES

3D bioprinting offers innovative solutions for fabricat-

ing complex biological structures by enabling precise layer-by-layer deposition of cell-laden bioinks (Hull et al., 2022). This technology, initially developed for tissue engineering and disease modelling, provides a more physiologically relevant environment compared to traditional 2D cultures, enhancing cell growth and interaction within an extracellular matrix-like framework (Jain et al., 2022; Xie et al., 2024b). In the context of MES, bioprinting can significantly enhance bioelectrochemical performance by constructing biofilms on electrodes with controlled architecture and composition (Lekshmi et al., 2023; da Rosa Braun et al., 2024). This approach leverages the ability to create highly organised microbial communities that optimise interactions between microbes and electrodes, improving electron transfer rates and metabolic activity (Wang et al., 2022; Wangpraseurt et al., 2022; Zhao et al., 2023). Therefore, we can achieve enhanced conductivity, surface area, and biocompatibility by utilizing bioprinting to fabricate bioanodes and biocathodes, ultimately leading to more efficient and effective MES.

4.1 Scaffold fabrication and bioink formulation

The success of bioprinting in MES applications depends heavily on the formulation of bioinks and the fabrication of scaffolds (Cui et al., 2022; Handral et al., 2022). Bioinks need to have appropriate rheological properties to ensure printability while maintaining cell viability and functionality (Schwab et al., 2020; Habib and Khoda, 2022). Common bioink materials include biocompatible hydrogels such as alginate, gelatin, and polyethylene glycol (PEG), which provide a supportive matrix for microbial cells (Cui et al., 2020; Mirek et al., 2022). Conductive materials like graphene or carbon nanotubes can be incorporated into the bioink to enhance the electrical properties of the printed structures (Zhou and Vijayavenkataraman, 2021; Moura et al., 2022).

Several 3D printing technologies are suitable for bioprinting, including direct ink writing (DIW), SLA, and laser-induced forward transfer (LIFT) (Zandrini et al., 2023). DIW is an extrusion-based method where bioink is extruded through a needle using pneumatic or mechanical force (Saadi et al., 2022). This technique allows for printing bioinks with high cell densities and viscosities, which is crucial for maintaining structural fidelity (Baier et al., 2022). However, the shear force exerted during extrusion can be harmful to cells, and the slow printing process can lead to cell dehydration and overexposure (Boularaoui et al., 2020; Adhikari et al., 2021; Taghizadeh et al., 2022). SLA and LIFT are laser-assisted, orifice-free printing methods. SLA uses a viscous photosensitive polymer solution that

solidifies upon exposure to light, enabling the construction of complex structures with high cell density and viability (Zheng et al., 2021; Zennifer et al., 2022). Despite its advantages, SLA can cause cell damage from UV light and photoinitiators and struggles with printing multiple resins or cells in a single structure (Quan et al., 2020; Xu et al., 2022a). LIFT, on the other hand, achieves high resolution and avoids clogging due to its nozzle-free design, but the rheological properties of the bioink also limit its application (Jentsch et al., 2021). For scaffold fabrication, porous structures are essential to support cell adhesion, growth, and nutrient diffusion (Zaeri et al., 2022; Flores-Jiménez et al., 2023). In scaffold-based models, hydrogels provide a 3D matrix that mimics the extracellular environment, maintaining cell viability and mechanical stability (Badekila et al., 2021; Tan et al., 2022). Natural hydrogels like agarose, alginate, and collagen offer inherent bioactivity but lack the mechanical strength required for long-term stability in MES (Pradhan et al., 2020; Radulescu et al., 2022). Synthetic hydrogels, such as PEG and polyacrylamide, provide tunable mechanical properties but are less biocompatible (Li et al., 2020; Nasution et al., 2022). To achieve a balance between biocompatibility and mechanical integrity, a blend of natural and synthetic hydrogels is often used (Kesharwani et al., 2021).

In general, extrusion-based 3D bioprinting is recommended for laboratory-scale trials in MES to create

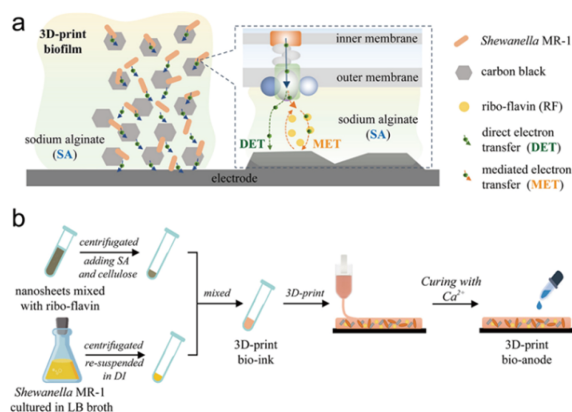
stable biofilms with high microbial load (He et al., 2021b; Wei et al., 2021). For industrial-scale applications, SLA or LIFT can be employed due to their ability to print thicker biofilms with complex internal structures, such as microchannels, that enhance mass transport and bioelectrochemical performance (Yi et al., 2021; Yu et al., 2023).

4.2 Current applications of bioprinting techniques in MES

Several studies have explored the diverse applications of 3D bioprinting in microbial electrochemistry, highlighting its potential to revolutionise this field. These studies can be categorised based on the 3D printing techniques employed, primarily focusing on extrusion bioprinting and electrochemical 3D printing (Fig. 3).

In extrusion bioprinting, bioink containing live microbes, hydrogels, and conductive materials is deposited through a nozzle to create 3D structures (Cui et al., 2020; Mobaraki et al., 2020). This method allows precise control over the spatial distribution of microbes and conductive materials, enhancing the functionality of bioelectrodes (Palmara et al., 2021; Zhao et al., 2023). One study investigated the use of 3D bioprinting to immobilise *Sporomusa ovata* cells on a cathode for microbial electrosynthesis, resulting in significantly increased acetate production compared to conventional methods (Krige et al., 2021). Another study enhanced

Extrusion bioprinting



Electropolymerization bioprinting

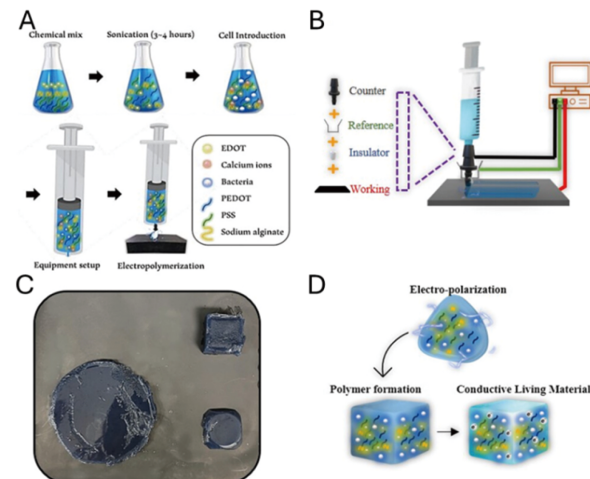


Fig. 3 Illustration of bioprinting techniques applied in MES. 1) Extrusion bioprinting for *Shewanella* bioanode with riboflavin-modified carbon black bioink: (a) EET in the designed 3D-bioprinted biofilm mimics, (b) Schematic of the preparation process, copyright 2023, American Chemical Society (Yang et al., 2024). 2) Electropolymerisation bioprinting for living exoelectrogens bioelectrodes: (A) 3D-printable ink preparation, (B) Electropolymerisation process design, visually representing the three-electrode system (including the counter electrode, work electrode, and reference electrode), (C) Polymer samples of the electropolymerised PEDOT building structure, (D) Electropolymerization stages of the EDOT monomer containing electrogenic bacteria, copyright 2023, John Wiley and Sons (Elhadad and Choi, 2023).

extracellular electron transfer efficiency within a 3D-printed *Shewanella oneidensis* MR-1 bioanode by incorporating riboflavin into the bioink as a redox mediator, leading to a significantly higher power density (Yang et al., 2024). Further research compared the performance of 3D bioprinted *Sporomusa ovata* cathodes on different metal meshes, finding that titanium was the most effective material due to its superior biocompatibility (Bajracharya et al., 2024). Additionally, the first 3D-printed living MFC anode using *Shewanella oneidensis* MR-1 cells was demonstrated, which showed promising performance and highlighted the potential of 3D bioprinting for large-scale MFC fabrication (Freyman et al., 2020).

Electrochemical 3D printing combines 3D printing with electropolymerisation to create biocompatible scaffolds with integrated microbes. This technique offers unique advantages for fabricating living bioelectrodes with tailored properties. A notable study introduced a method for fabricating living bioelectrodes using electrochemical 3D printing, where the bioelectrodes consisted of a poly 3,4-ethylene dioxythiophene scaffold embedded with exoelectrogenic bacteria, enabling efficient electron transfer and potential applications in bioelectronics (Elhadad and Choi, 2023).

These studies provide compelling evidence of the transformative potential of 3D bioprinting in microbial electrochemistry. Extrusion bioprinting has demonstrated its ability to enhance MES performance by improving biofilm stability, mass transfer, and electron transfer efficiency. Meanwhile, electrochemical 3D printing has emerged as a promising technique for creating biocompatible scaffolds with integrated microbes, opening doors for novel bioelectronic applications. As 3D bioprinting technology continues to advance, we can expect even more innovative and effective applications in microbial electrochemistry. These advancements hold immense promise for revolutionising sustainable energy production, environmental remediation, and biosensing technologies. In addition to the advantages above, 3D bioprinting offers improved biocompatibility, enhanced mass transfer, tailored electrode design, and versatility, making it a pivotal tool in the development of next-generation microbial electrochemical systems.

5 Current challenges and opportunities

While the integration of 3D printing in MES holds

significant promise, several challenges and opportunities merit consideration. One primary challenge is the optimisation of printing parameters to ensure the precise fabrication of electrode architectures conducive to microbial adhesion and electron transfer (Ratheesh et al., 2021). Achieving optimal porosity, surface roughness, and geometry while maintaining structural integrity remains a complex task. Moreover, the selection and formulation of bioinks for bioprinting applications require careful consideration of rheological properties, biocompatibility, and conductivity to support cell viability and electrochemical activity effectively. Another challenge lies in scaling up 3D printing techniques for industrial-level MES applications (Boas et al., 2022; Lekshmi et al., 2023). While extrusion-based printing techniques are suitable for laboratory-scale trials, they may not be cost-effective or efficient for large-scale manufacturing. Implementing scalable printing methods such as SLA or LIFT requires addressing technical and logistical hurdles to ensure reproducibility, consistency, and cost-effectiveness (Ngo et al., 2018; Fidan et al., 2023). Furthermore, the long-term stability and durability of 3D-printed electrodes in harsh MES environments remain a concern (Choi, 2022; Badreldin et al., 2024). Electrode materials and printing techniques must be carefully chosen to withstand mechanical, chemical, and biological stresses over extended operational periods. Additionally, the integration of sensors and monitoring devices within 3D-printed MES components presents both challenges and opportunities for real-time process control and optimisation (Choi, 2022; Li et al., 2024).

Further research must be done to understand how electrode morphology improves performance. How much do we need to increase our surface area? Guo et al. (2020) found that their 3D-printed anodes resulted in high coulombic efficiencies but low current densities. They blamed the enlarged surface-area/ reactor-volume ratios, as well as the heterogeneity of the biofilm. New research should focus not just on making larger surface area electrodes, but on actually integrating them with reactors of the right size. A good proportion between electrode surface area and electrolyte volume will ensure that this area can be colonised with microbes that have enough nutrients and electron donors/acceptors. Moreover, studies on the morphology and disposition of the biofilm on the electrode are also lacking. It is crucial to understand the interaction between electrode and biofilm. Combining methods such as Scanning Electron Microscopy, Confocal Microscopy, or 16s rRNA sequencing can give us insights into the type of biofilm, distribution, or

community. However, this should be done thoroughly, looking into different areas of the electrode's surface, inside and outside, and considering different external variables, such as current, electrolyte, or inoculum of choice. Another important question is whether we need a larger surface area in terms of projected area or volume occupied. Even though some studies see an improvement in performance when using 3D electrodes, the actual use of the inside of the electrode is not well known. The availability of nutrients, clogging of pores, and others need to be assessed. For that, more and long-term experiments should be run with these 3D electrodes.

On another note, sustainability studies such as life cycle assessments need to be performed, not only for new 3D-printed electrodes and reactor bodies but also for more traditional BES materials. There is a knowledge gap in the sustainability challenges these technologies face, and some of the proposed 3D-printed parts and methods might have a more, or less environmental impact than typical materials when looking into a life cycle assessment (Aguilera Flores et al., 2024).

Despite these challenges, numerous opportunities exist to advance the application of 3D printing in MES manufacturing. The ongoing development of novel materials, printing techniques, and bioink formulations holds promise for overcoming existing limitations and enhancing the performance and functionality of 3D-printed electrodes and reactor components. Besides, 3D printing has opened up multiple new applications within MES, such as wearable MFCs. Integrating non-toxic, storable, spore-forming bacterial cells into a flexible, disposable paper-based MFC platform offers a novel approach to energizing single-use wearable devices powered by sweat (Gao et al., 2022). In this case, collaborative interdisciplinary research efforts between material scientists, bioengineers, electrochemists, and environmental engineers are essential for driving innovation in this field.

From an economic perspective, a techno-economic assessment must be done to assess the contribution to capital and operating costs when using 3D-printed electrodes or 3D-printed parts in bioelectrochemical systems. Jourdin et al. (2020) studied 3 different designs for integrated systems containing a microbial electrosynthesis step. They found that anodes are the main CAPEX while electricity usage is the main OPEX. 3D printing could portray opportunities to make more efficient anodes and host thicker biofilms that will provide higher current densities. However, inks and further post-printing treatments are still expensive.

Efforts should be made to find 3D-printing materials that are low-cost, durable, easily disposable, and, ideally, from revalorised waste streams that are also biocompatible and have good electrochemical properties. (Chung and Dhar, 2021).

6 Conclusions

MES plays a vital role in wastewater treatment and resource recovery. The integration of 3D printing technology revolutionises MES design by allowing precise fabrication of reactor components to enhance fluid dynamics and electron transfer. This advancement accelerates reactor development, reduces costs, and facilitates iterative improvements for commercialisation. Tailored ink materials optimise the conductivity and biocompatibility of bioelectrodes, enhancing energy generation and microbial-electrode interactions. Bioprinting enables precise biofilm deposition on electrodes, improving electron transfer rates. Overall, 3D printing enhances electrode design in MES, boosting performance and operational efficiency for sustainable wastewater treatment and resource recovery.

Abbreviation List

ABS	Acrylonitrile butadiene styrene
DIW	Direct ink writing
EIS	Electrochemical impedance spectroscopy
FDM	Fused deposition modeling
FFF	Fused filament fabrication
FTO	Fluorine-doped tin oxide
GA	Graphene aerogel
GO	Graphene oxide
ITO	Indium tin oxide
LDM	Liquid deposition modeling
LIFT	Laser-induced forward transfer
L-PBF	Laser-powder bed fusion
MES	Microbial electrochemical system
MFC	Microbial fuel cell
NZVI	Nano zero-valent iron
ORR	Oxygen reduction reaction
PEG	Polyethylene glycol
PETG	Polyethylene terephthalate glycol
PLA	Poly(lactic acid)
PPy	Polypyrrole

PyrC	Pyrolytic carbon
SLA	Stereolithography
SLM	Selective laser melting

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