frontiers
in Energy Research

A Mini-Review on Applications of 3D Printing for Microbial Electrochemical Technologies

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For the past two decades, many successful applications of microbial electrochemical technologies (METs), such as bioenergy generation, environmental monitoring, resource recovery, and platform chemicals production, have been demonstrated. Despite these tremendous potentials, the scaling-up and commercialization of METs are still quite challenging. Depending on target applications, common challenges may include expensive and tedious fabrication processes, prolonged start-up times, complex design requirements and their scalability for large-scale systems. Incorporating the three-dimensional printing (3DP) technologies have recently emerged as an effective and highly promising method for fabricating METs to demonstrate power generation and biosensing at the bench scale. Notably, low-cost and rapid fabrication of complex and miniaturized designs of METs was achieved, which is not feasible using the traditional methods. Utilizing 3DP showed tremendous potentials to aid the optimization of functional large-scale METs, which are essential for scaling-up purposes. Moreover, 3D-printed bioanode could provide rapid start-up in the current generation from METs without any time lags. Despite numerous review articles published on different scientific and applied aspects of METs, as per the authors' knowledge, no published review articles explicitly highlighted the applicability and potential of 3DP for developing METs. Hence, this review targets to provide a current overview and status of 3DP applications for advancing METs and their future outlook.

Keywords: 3D printing, additive manufacturing, microbial electrochemical technologies (MET), microbial electrochemical cells (MXCs), microbial fuel cells (MFCs)

INTRODUCTION

Microbial electrochemical technologies (METs) are unique platforms that combine microbial metabolism with electrochemistry for various value-added applications (Sravan et al., 2021). Over the last 2 decades, many different applications of METs have been demonstrated: 1) bioenergy generation, such as bio-electricity in microbial fuel cells (MFCs) (Munoz-Cupa et al., 2020; Sravan et al., 2021), bio-hydrogen in microbial electrolysis cells (MECs) (Hua et al., 2019; Rousseau et al., 2020), and bio-methane in MEC assisted anaerobic digesters (Zakaria and Dhar, 2019; Huang et al., 2020); 2) synthesis of platform chemicals, such as hydrogen peroxide (Chung et al., 2020b; Zhao et al., 2021); 3) nutrient recovery (Zou et al., 2017; Barua et al., 2019); 4) water desalination (Al-Mamun et al., 2018; Jafary et al., 2020); 5) biosensors (Do et al., 2020; Chung et al., 2020a); and many more. Despite such tremendous potential, studies emphasized the challenges in system design and fabrication, which must be addressed to improve their performance and

TABLE 1 | Summary of studies used 3D printing for the fabrication of key components in microbial fuel cells.

Component	Material/ink	Application	Major findings/remarks	References
Reactor body	Polycarbonate acrylonitrile butadiene styrene	Biosensor	Rapid detection (COD: 2.8 min; cadmium: 12 min) and high sensitivity (COD: 3–164 ppm; cadmium: 1–50 µg/L)	Di Lorenzo et al. (2014)
Reactor body	Polycarbonate acrylonitrile butadiene styrene	Biosensor	Rapid detection (40 min) of COD (25–200 mg/L)	López-Hincapié et al. (2020)
Reactor body	Acrylic based photosensitive polymer	Biosensor	This study focused on understanding performance of biosensor using anode materials with different morphologies	Quaglio et al. (2019)
Reactor body	Nanocure® resin	Power generation	The primary objective was to develop a liquid-handling robot for automated feeding of substrate to 3D printed MFCs	Ieropoulos et al. (2016)
Anode	UV curable resin (post-treated via carbonization)	Power generation	Compared to carbon cloth anode, 3D printed porous carbon anode improved maximum voltage generation by ~2.4 times	Bian et al. (2018a)
Anode	UV curable resin (post-treated with copper for surface modification)	Power generation	3D printed electrode produced 3-fold higher maximum voltage generation than carbon cloth electrode	Bian et al. (2018b)
Anode	Graphene oxide, ferric ions, and magnetite nanoparticles	Power generation	3D printed electrode produced 7.9-fold higher volumetric current density than carbon felt electrode	He et al. (2021)
Anode, cathode, and chassis	PLA (post-treated with graphite or nickel powder coating)	Power generation	Carbon-coated 3D printed PLA anode showed power generation comparable to modified carbon veil; however, 3D printed PLA cathodes underperformed regardless of the coating	You et al. (2020)
Anode (dual- and single-chamber) cathode (dual-chamber only)	PLA	Power generation	3D printing provided precise manufacturing of electrodes (9.7 cm^2) with lower costs	Jannelli et al. (2018)
Anode	PLA	Power generation	3D printed anode showed 4.1-times lower power generation than conventional plain carbon veil anode	You et al. (2017)
Anode	Stainless steel and titanium alloy (post-treated with polyaniline coating)	Power generation	Electrode with unique microporous skeleton structure was fabricated with 3D printing, which is impossible for traditional manufacturing methods; polyaniline coating further developed the performance of 3D printed electrodes	Zhou et al. (2017)
Anode	Aluminum alloy (AlSi10 Mg)	Power generation	3D microporous electrode could produce energy recovery up to ~3 kWh/m^3 per day	Calignano et al. (2015)
Cathode	Alginate + activated carbon	Power generation	Cost-effective and rapid fabrication of electrode	Theodosiou et al. (2020)
Bioanode	Sodium, alginate, sigma, cellulose, *Shewanella oneidensis* MR-1	Power generation	First demonstration of 3D printed electroactive bacterial bioanode	Freyman et al. (2020)
Membrane	Tangoplus and natural rubber latex	Power generation	The 3D printed latex membrane showed comparable power density as commercial CEM, while 3D printed tangoplus membrane was more resistant to biofouling. The fabrication costs of both membranes were lower than commercial CEM	Philamore et al. (2015)
Membrane	Porous filaments of rubber-elastomeric polymers	Power generation	3D printed polymeric membranes showed 1.4-fold higher power generation than a commercial cation exchange membrane; however, fabrication cost was higher	You et al. (2017)
Membrane electrode assembly (MEA)	Fimo™ air-dry clay and terracotta air-dry clay	Power generation	Air-dry clay-based 3D printed MEAs could produce ~50% more power than the controls (commercial CEM and kilned red terracotta). Moreover, the fabrication cost of 3D printed MEAs was cheaper than commercial CEM-based MEA	Theodosiou et al. (2019)

robustness, especially for scaling-up and commercialization (Dhar et al., 2016b; Sim et al., 2018; Zakaria and Dhar, 2019). Specifically, developing a low-cost and efficient fabrication technique is imperative for advancing METs (Bian et al., 2018a; Theodosiou et al., 2020; You et al., 2020). The utilization of conventional subtractive manufacturing methods is time-consuming and highly laborious and can generate wastes from cutting materials away from larger pieces (Theodosiou et al., 2020; You et al., 2020). Moreover, the precise manufacturing of miniaturized METs for applications like biosensors is also challenging. For instance, construction defects such as widening of spacings between parts and fittings or unwanted pinholes may result in the intrusion of microbubbles, which can significantly reduce the biosensing performance (Fraiwan et al., 2013). Therefore, developing a highly functional, low-cost, and environmentally sustainable fabrication method is essential for reducing time lags toward commercialization of METs (You et al., 2017; Bian et al., 2018a; You et al., 2020).

The additive manufacturing (AM) process or three-dimensional printing (3DP) has been expanding rapidly in recent years. Briefly, 3DP generates 3D structures from computer-aided design (CAD) models by adding the material layer-by-layer, allowing rapid and precise fabrication of sophisticated structures and devices with complex geometry with minimum human interventions (Geissler and Xia, 2004; Bian et al., 2018b; You et al., 2020). Due to these promising features, 3DP has been implemented in various fields, including industrial prototype printing, aerospace (Griffiths, 2015), medical implants (Murphy and Atala, 2014; Rasperini et al., 2015), and arts (Walters and Davies, 2010). Moreover, 3DP has been applied to various energy-generating systems, such as solar cells (Angmo et al., 2013; Vak et al., 2015) and batteries (Sun et al., 2013; Izumi et al., 2014). Owing to the increased popularity and applicability, 3DP has recently been applied to fabricate different parts of METs, such as reactor bodies, electrodes, and membranes (**Table 1**). To the authors' knowledge, no review articles critically appraised the literature published on the application of 3DP for developing METs. Hence, this paper aims to provide a critical appraisal of the studies that utilized 3DP for METs. Furthermore, potential future developments and recommendations are outlined.

APPLICATION OF 3D PRINTING FOR MICROBIAL ELECTROCHEMICAL TECHNOLOGIES

Reactor Body

The majority of METs are primarily developed for water/wastewater applications; thus, different types of reactor architectures can significantly influence the effective flow and hydrodynamics (e.g., mass transfer within biofilms) and thereby influence their performance (Kim et al., 2014; Jiang et al., 2015; Yi et al., 2020). Hence, optimizing the shape of the system, working volumes, fluid inlet/outlet settings are critical (Logan et al., 2015; Massaglia et al., 2017; Yi et al., 2020). For instance, a drop-like-shaped MFC could perform better than a square-shaped one due to the larger effective exposed area (Massaglia et al., 2017). Moreover, the addition of baffles in the anode chamber could result in higher voltage output in MFCs due to reduced dead zones where the flow velocity is <5% of the maximum velocity (Yi et al., 2020). However, fabrication of such designs and configurations can be extremely challenging using the traditional manufacturing technique, especially for the miniaturized METs. Several studies have reported successful utilization of 3DP for precise fabrication of reactor bodies for METs (see **Table 1**).

Di Lorenzo et al. (2014) applied 3DP for rapid fabrication of a miniaturized (2 cm^3) air-cathode MFC biosensor. Their fabricated MFC biosensor exhibited a good detection range (3–164 ppm), high sensitivity (0.5 μA mM^{-1} cm^{-2}), a fast response time (2.8 min), and fast recovery time (12 min) while detecting cadmium ions in water samples. Moreover, Quaglio et al. (2019) developed 3D-printed a MFC biosensor (12.5 ml), which allowed optimizing hydrodynamics (optimal fluid motion)

and morphology of anode electrode (porosity) via computational fluid dynamics (CFD) modeling for quantification of sodium acetate in water samples. Although 3DP can allow bulk production of small-scale reactors for applications like biosensing within reasonable timeframes and costs, direct 3D printing of large-scale systems is not yet feasible. Nonetheless, approaches like combining 3DP and CFD modeling can enable low-cost advanced reactor engineering for large-scale field applications (e.g., wastewater treatment) of METs. For instance, governing equations for mass, momentum, and energy conservation to find numerical solutions in CFD modeling are not scale-dependent (Parra-Cabrera et al., 2018). Thus, when combined with CFD, 3D printed lab-scale prototypes can still be used for the design and optimization of large-scale systems.

Electrodes

The development of electrodes that possess highly porous structures for optimal bacterial adhesion and excellent electrochemical performance for high current output has been of great interest to the MET research communities (Zhou et al., 2017; Bian et al., 2018a; Bian et al., 2018b). Notably, complex macro-porous 3D electrode materials have been found suitable for providing a larger surface area for the biofilm developments and thereby improve the electrochemical performance of METs (Hindatu et al., 2017; Zhou et al., 2017). However, the major hurdle is in the precise construction of electrodes with geometrical complexities (e.g., porous electrodes) using the traditional manufacturing method (Zhou et al., 2017; Bian et al., 2018a; Bian et al., 2018b). Recently, researchers have explored 3DP for rapid prototyping of such ideal electrodes METs.

Zhou et al. (2017) reported that 3DP could allow the precise design of porous 3D structured X-shape skeleton electrodes with titanium and stainless steel as printing materials. The further modification of 3D-printed metal electrodes with polyaniline could provide a crumpled and biocompatible surface. Notably, the polyaniline coating of the 3D-printed titanium electrode could increase power density up to 400 times than the uncoated one. Calignano et al. (2015) developed an aluminum alloy (AlSi10 Mg) anode using 3DP, which could enable energy output of up to 3 kWh/m^3 per day from an MFC. The authors suggested that the 3D microporous coral skeletal structure could provide surface roughness and excellent biocompatibility to electroactive microbes. However, metal 3D printing is still extremely expensive (Pumera, 2019). Moreover, some metal alloys used for 3DP (e.g., AlSi10 Mg) alloy are prone to corrosion (You et al., 2017; Revilla et al., 2020). Previous studies also demonstrated the feasibility of utilizing polymeric materials, such as polylactic acid (PLA), UV curable resin, for 3D printing of electrodes for METs (see **Table 1**). However, the electrochemical performance of 3D-printed polymeric electrodes is not always satisfactory. For instance, power generation from the PLA electrode developed by You et al. (2017) was ~4 times lower than the control (plain carbon veil) with the same geometric structures. Despite conductive properties, thin protective polymer layers on the surface of 3D-printed

polymeric electrodes can restrict electrochemical performance, which can be removed by surface modification (Pumera, 2019). Bian et al. (2018a) proposed carbonization of the 3D-printed anode from polymer resin to enhance the biocompatibility, which could produce significantly higher power density than a traditional carbon cloth anode (233.5 vs. 69 mW/m^2). Similarly, surface modification of a 3D-printed polymeric electrode with copper could produce 12.3-fold higher power than copper mesh electrodes (Bian et al., 2018b). Most recently, He et al. (2021) developed a 3D-printed graphene oxide (GO) aerogel anode with a customized printing ink combining GO, ferric ions, and magnetite nanoparticles. The hierarchical pores in their GO aerogel electrode could provide effective mass transfer of substrates, leading to 7.9 folds higher volumetric current output than carbon felt anode. Thus, utilizing 3DP for electrode fabrication has shown promising results. While cost and performance are often the primary aspects considered by researchers, ensuring the long-term stability (chemical stability, corrosion resistance, etc.) of 3D-printed electrodes would be critical.

Bioelectrode

Besides electrodes and other physical components of METs, electroactive microorganisms are vital components affecting METs performance (Koch and Harnisch, 2016). The selection and enrichment of kinetically efficient electroactive bacteria (EAB) are often challenging (Torres et al., 2009; Dhar et al., 2016a). Due to slow growth kinetics, the time required for complete enrichment of anode or cathode biofilms can be significant (e.g., months-years), ultimately prolonging the reactor start-up time (Zakaria and Dhar, 2019). Several studies have utilized the effluent from MET reactors as an inoculum source (Zakaria et al., 2018; Barua et al., 2019; Chung et al., 2020a), which can expedite the enrichment process of electroactive biofilms. However, an innovative approach for the rapid establishment of electroactive biofilms is still required. A recent study by Freyman et al. (2020) suggested that 3DP can provide an attractive solution to such time-consuming biofilms development procedures. They investigated 3DP for constructing a bioanode with *Shewanella oneidensis* MR-1 as living inks. More importantly, *Shewanella oneidensis* MR-1 survived during the 3D printing process, and their MFC produced a stable current for almost 93 h. Of note, prior to their study, 3DP was applied to engineer 3D objects and structures with microorganisms, such as *Escherichia coli*, *Pseudomonas putida*, *Acetobacter xylinum*, yeast cells (Lehner et al., 2017; Schaffner et al., 2017; Qian et al., 2019). Particularly, due to the adhesion of cells during the process, objects 3D-printed with microbes could potentially achieve a high cell density. Nonetheless, based on the authors' knowledge, Freyman et al. (2020) first demonstrated the feasibility of 3D-printed bioanode, potentially opening up new possibilities for high-performance METs.

Ion-Exchange Membranes

Ion-exchange membranes (IEM) are imperative, especially for the dual-chamber METs, to facilitate the ion transport between anode and cathode, governing the electroneutrality (Dhar and Lee, 2013; Leong et al., 2013). The IEMs can alter the current output, depending on their relative surface area, which is associated with ionic conductivity and internal resistance of METs (Zuo et al., 2007; Philamore et al., 2015). Hence, increasing the specific ion-exchange area of IEM is great of interest, but it is often a major challenge (Philamore et al., 2015). Notably, commercially available IEMs can be costly (e.g., >700 USD/m^2 for Nafion) due to the complex manufacturing process (Yee et al., 2012; Dhar and Lee, 2013). Besides, IEMs require pre-treatment processes (Ghasemi et al., 2013; Rahimnejad et al., 2014), which can be time-consuming. Therefore, an efficient and low-cost membrane manufacturing method is greatly needed.

Several studies implemented 3DP as an alternative fabrication method for IEMs (Philamore et al., 2015; You et al., 2017; Theodosiou et al., 2019). Philamore et al. (2015) have manufactured IEMs for METs utilizing 3DP at a much lower cost than commercial CEM ($0.00112–0.00357 for latex and $0.16 for Tangoplus resin vs. $0.22–40 for commercial CEM, per 20 cm^2, USD). The power generation from an MFC with a 3D-printed latex membrane was comparable to commercial CEM (10.51 vs. 11.39 μW). In contrast, maximum power generation was much lower in the MFC with a 3D-printed Tangoplus membrane (0.92 μW). Nonetheless, the latex membrane was more prone to fouling and almost damaged after 210 days of operation, while the Tangoplus membrane was more resistant to biofouling. In contrast, 3D printed membrane using rubber-elastomeric polymers developed by You et al. (2017) could increase maximum power output up to 1.4 times than commercial CEM; however, the fabrication cost of the 3D-printed membrane was higher than commercial CEM. Thus, these results suggested that selecting low-cost and durable polymeric materials would be necessary for fabricating economic and technically sustainable 3D-printed IEMs. However, previous studies primarily focused on the performance of 3D-printed IEMs in terms of current output and biofouling potential, while limited information provided on other critical features, such as proton permeability, membrane resistance, substrate loss, and oxygen diffusion (in MFCs) (Dhar and Lee, 2013).

Membrane Electrode Assembly (MEA)

The spacing between electrodes is critical for minimizing internal energy losses in METs (Moon et al., 2015b). Therefore, MEA (membrane sandwiched between electrodes) has been considered by many researchers to reduce internal energy losses and thereby improve the performance of METs (Moon et al., 2015b). Typically, MEAs are prepared by pressing membrane and electrodes with or without heat treatment (Theodosiou et al., 2019), which has multiple drawbacks. For instance, a high degree of hydrophilicity can often cause anolyte leakage from the surface of MEA (Mashkour et al., 2021). Furthermore, uneven physical contact between membrane and electrode resulting from the conventional fabrication method can increase internal resistance and ultimately lower the current output (Moon et al., 2015a; Vilela et al., 2020). Therefore, precise fabrication methods for MEA are required. Recently, Theodosiou et al. (2019) compared 3D-printed MEAs with a commercial CEM.

The 3D-printed MEAs from air-dry clay materials exhibited up to 50% higher power output (130 vs. 66 μW) for >70 days and provided 34–51 times cheaper production cost. Although the results were promising, future studies should focus on a comprehensive investigation of underlying mechanisms that can provide a superior performance of 3D-printed MEAs over the conventional ones.
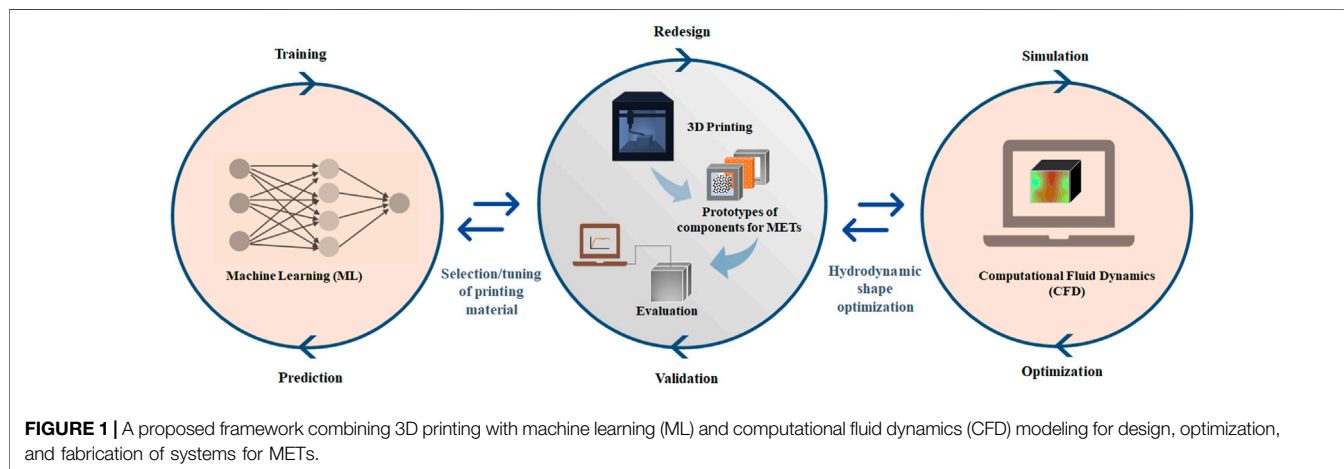
SUMMARY AND OUTLOOK

With the recent advancement in 3D printing technologies, their application in METs has been gaining attention. Previous studies demonstrated the potential as a precise and rapid fabrication method for different components of METs. Notably, 3DP could control complex geometric structures of electrodes with customized properties (e.g., porosity, roughness, etc.) to improve efficiency over traditional electrodes (discussed earlier). Although outcomes have been promising, several drawbacks, such as high cost, low durability, biocompatibility, and electrochemical properties of 3D printed components (Angmo et al., 2013; Wei et al., 2015; You et al., 2017), may limit the application of 3DP for METs (Bian et al., 2018a). Therefore, future research should continue to seek low-cost but well-performing 3D-printable materials for MET components.

While polymeric materials would be a better choice for keeping the cost low (You et al., 2017), activation or post-treatment would be necessary to achieve desired performance from 3D-printed electrodes (Baran and Erbil, 2019; Pumera, 2019). It is expected that activation steps will increase the fabrication time and cost. Recently, various simple and low-cost activation methods for 3D-printed polymer electrodes have been developed for improving conductive properties for various electrochemical applications (Browne et al., 2018; Pumera, 2019). For instance, dimethylformamide (DMF) immersion and electrochemical activation by applying constant potential have recently been demonstrated as simple and cost-effective methods for activating 3D printed polymer/graphene by removing the insulating polymer layer (Browne et al., 2018; Pumera, 2019). These methods should be explored

for developing highly conductive 3D-printed polymer electrodes for METs. Moreover, strategies for improving biocompatibility would be an additional aspect that needs to be addressed by the METs research community. Likewise, there have been significant developments toward developing advanced 3D-printed membranes for various industrial applications (Lee et al., 2016; Lv et al., 2017). Thus, multidisciplinary collaborations in these areas can promote rapid development.

As an emerging concept, the feasibility of 3D-printed bioelectrode could still be questionable. However, EAB can rapidly recover from extreme and harsh environments, such as extremely low-temperature settings below freezing point (Winfield et al., 2015; Cho et al., 2020), long-term (5–10 days) starvation (Ruiz et al., 2015; Dhar et al., 2018). The digital printing of electroactive cyanobacterium onto the paper electrode was also successful in the past (Sawa et al., 2017). Thus, the concept of 3D-printed bioelectrode should be highly feasible with further developments, which can undoubtedly shorten the start-up time of METs. Additionally, there are still many other possibilities to utilize such an innovative approach to further advance METs. In METs, energy recovery from complex organic substrates requires a partnership between EAB and their syntrophic partners (e.g., fermentative bacteria), as complex substrates (e.g., fermentable organics) can not be directly oxidized by EAB (Parameswaran et al., 2009; Dhar et al., 2019; Zakaria and Dhar, 2020). However, existing literature provides limited information on how to manipulate or develop a well-balanced syntrophic community for high-performance METs. It can be envisioned that the application of 3DP can offer a pragmatic solution to develop living bioelectrode with a designer syntrophic co-culture with target functionality (e.g., electron recovery from a specific complex electron donor). Thus, it will be interesting to see the feasibility and sustainability (i.e., long-term reliable operation) of such new opportunities.

Moreover, the broader impact of 3DP can be facilitated by incorporating other research tools, such as CFD and machine learning (ML) modeling tools (**Figure 1**). As discussed earlier, the combination of CFD and 3DP bears immense potential as a design optimization tool for large-scale METs. Particularly, the

FIGURE 1 | A proposed framework combining 3D printing with machine learning (ML) and computational fluid dynamics (CFD) modeling for design, optimization, and fabrication of systems for METs.

CFD model can provide critical information on the crucial parameters, such as maximum flow locations and dead zones with different designs for METs (Kim et al., 2014; Yi et al., 2020). The information can be adopted to rethink and redesign METs that can be rapidly prototyped with 3DP and tested for further validation. Moreover, incorporating ML techniques with 3DP can also be useful. For instance, previous studies substantiated the significance of printing inks on the performance and sustainability of 3D-printed components of METs (Philamore et al., 2015; You et al., 2017; Theodosiou et al., 2019). ML can be utilized to select efficient printing materials for achieving specific characteristics, such as conductivity, porosity, and surface roughness required for 3D-printed components of METs. The lack of METs-specific training data and data source variability could be barriers to such developments. However, there have been developments toward ML algorithm validation with a small amount of training data (Vabalas et al., 2019). Therefore, future studies should explore these options for optimal design and fabrication of 3D-printed components, especially for larger-scale METs, in which, the current challenges and limitations were discussed earlier.

AUTHOR CONTRIBUTIONS

TC: Conceptualization, Investigation, Writing-original draft, Writing-review & editing BD: Conceptualization, Funding acquisition, Supervision, Writing-original draft, Writing-review & editing.

ACKNOWLEDGMENTS

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