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REVIEW

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From bibliometric analysis: 3D printing design strategies and battery applications with a focus on zinc-ion batteries

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Abstract

Three-dimensional (3D) printing has the potential to revolutionize the way energy storage devices are designed and manufactured. In this paper, we explore the use of 3D printing in the design and production of energy storage devices, especially zinc-ion batteries (ZIBs) and examine its potential advantages over traditional manufacturing methods. 3D printing could significantly improve the customization of ZIBs, making it a promising strategy for the future of energy storage. In particular, 3D printing allows for the creation of complex, customized geometries, and designs that can optimize the energy density, power density, and overall performance of batteries. Simultaneously, we discuss and compare the impact of 3D printing design strategies based on different configurations of film, interdigitation, and framework on energy storage devices with a focus on ZIBs. Additionally, 3D printing enables the rapid prototyping and production of batteries, reducing leading times and costs compared with traditional manufacturing methods. However, there are also challenges and limitations to consider, such as the need for further development of suitable 3D printing materials and processes for energy storage applications.

KEYWORDS

3D printing, battery commercialization, bibliometric analysis, electrode configurations, zinc-ion battery

1 | INTRODUCTION

Energy storage devices, batteries in particular, are important to sustainable development because they enable the use of renewable energy sources, reduce greenhouse gas emissions, improve the reliability and stability of the grid, and reduce energy costs.[1–4] As the demand for clean and reliable energy continues to grow, the importance of energy storage devices and environmental technology is only likely to increase in the coming years.[5–8] There is currently a significant amount of research and development taking place in the field of energy storage devices.[9] One of the main areas of focus is the development of more efficient, safe, flexible, and cost-effective batteries. Researchers are exploring new materials, manufacturing methods, and designs to achieve the high demands of next-generation batteries.[10] The development of energy storage technologies, based on monovalent cations such as Li ions, Na ions, K ions, and multivalent cations such as Zn ions, Mg ions, and other energy storage technologies, has been driven by the need for more efficient and cost-effective devices.[11–19] As for monovalent cation batteries, Li-ion batteries (LIBs), which are currently the most widely used type of battery, have undergone significant development in recent years, with improvements in energy density, lifespan, and safety. Na-ion batteries and K-ion batteries are newer technologies that are currently being developed as potential alternatives to LIBs.[20–22] Among all candidates for multivalent cation batteries, numerous researchers have recently become interested in Zn-ion batteries (ZIBs) due to their numerous advantages over other types of batteries.[23] In the future years, ZIBs has the potential to play a significant role in the transition to a more sustainable energy mix. Zn is a relatively inexpensive and abundant element, making it a far less expensive raw material than Li, which is used in traditional LIBs.[24–27] It indicates that ZIBs might be manufactured at a significantly cheaper cost, making them more accessible to a broader variety of consumers. The high energy density of ZIBs is a second advantage over most other energy storage systems.[28] The comparatively high energy density of ZIBs allows them to store large amounts of energy in a relatively small volume.[29–31] This makes them have the potential for applications where size and weight are crucial, such as portable electronics. Additionally, ZIBs have a lengthy lifespan. These batteries can maintain a high level of performance across a large number of charge and discharge cycles.[32] This makes them a cost-effective option for battery-intensive applications. Importantly, aqueous ZIBs are safer than conventional LIBs.[33] Under specific conditions, such as damage or overheating, LIBs are known to catch fire or explode. The absence of these safety issues makes aqueous ZIBs a safer option for a range of applications. Finally, ZIBs are less harmful to the environment. The production and disposal of typical LIBs have a detrimental impact on the environment due to the usage of toxic chemicals and the difficulty of recycling the batteries. In contrast, ZIBs are created from nontoxic substances and are easily recyclable, making them a more sustainable option.[34]

In the last few years, novel electrode materials and electrolytes have been explored and new manufacturing methods have been developed to improve the electrochemical performance of ZIBs.[35–37] The rate of the redox reaction at the electrode, the transport rate of Zn ions, and the electron conductivity are very important factors to improve the performance of energy storage devices.[38] ZIBs use Zn or Zn-containing materials as the anode and manganese dioxide, vanadium oxide, Prussian blue, and other materials as the cathode.[39–41] For ZIBs, ion intercalation and dendrite growth have become an obstacle to development. In recent years, studies have shown that thin films as a barrier film option can have a controlled regulation toward Zn ion deposition, and organic functional groups may modulate Zn ion transport to achieve a uniform electric field and form a flat, dendrite-free Zn metal precipitation layer, enhancing the efficient cycling stability of ZIBs.[42] The benefits of incorporating a thin film process will not only enhance the efficiency of the battery but also provide the battery the flexibility and wearable properties to accommodate the development of wearable and portable electronic devices.[43] As for thin-film electrodes, Zn ions can be easily transferred to the electrode, and the increased efficiency of the Zn ion transport facilitates a sufficient reaction between the active material in the electrode and the Zn ions in the electrolyte. In fact, not all components of the battery have the capacity to release or store electrical energy. Some components do not have the chemical reaction conditions or the chemical reactions that occur have an impact on the oxidation or reduction reactions at the electrodes regarding the storage of energy, and these components can significantly reduce the energy or power density of the battery. Therefore, it is essential to create high-energy density energy storage devices. Although increasing electrode thickness can increase the current strength to some extent, the energy density of the cell decreases as the electrode thickness increases. The thickness of the electrodes also affects the distance and resistance of the electron transfer, and the increased resistance and distance of electron transfer will also lead to a decrease in the efficiency of the cell.[44] The significant increase in cell size and the relatively small reaction area can also lead to uncontrollable growth of dendrites, which can threaten cell safety and stability. Three-dimensional (3D) porous structures may offer a new method to solve these problems. On the one hand, 3D structures can reduce ion transport distances, and on the other hand, porous structures can improve the availability of electrodes. The porous structure can also reduce the current uneven distribution on the anode surface to alleviate the growth of dendrites, avoiding dendrites from piercing through the separator, and reducing battery life.[45] As for complex structures such as 3D porous structures, traditional chemical processes are difficult to achieve with precision. Additive manufacturing under precise computer control, also known as 3D printing, is well suited for such precise work in the manufacture of porous structures.

Several 3D printing technologies have been reported recently, 3D printing technology is being used substantially to produce finely detailed components due to its high level of flexibility and precision.[46] In principle, 3D printing is the stacking of materials (inks) to obtain the target object by curing methods.[47] It is a process of continually adding materials: a model is first created in a computer, and then the raw materials are stacked precisely and accurately under computer control so that the finished print can have any shape characteristic depending on the modeling in the computer. In a sense, the finished 3D printed product results from many layers of print stacked on top of each other. This means that the 3D printer is printing in successive layers, forming a layer of the printed surface, such as traditional printing, hence the so-called "print." Because of the flexibility and precision of 3D printing, this technology has an extensive range of applications, and almost all fields will be involved in the application of this technology.[48] And 3D printing is used chiefly in the manufacture of delicate parts. One potential application of 3D printing in energy storage devices is the production of battery electrodes.[49] Traditional battery manufacturing methods often involve the use of expensive and time-consuming processes, such as roll-to-roll printing and electroplating. 3D printing allows for the rapid production of electrodes with complex shapes and structures, which can potentially improve the performance of the battery.[50] Another potential application of 3D printing in energy storage devices is the production of custom-shaped batteries.[51] Traditional battery shapes are often dictated by the manufacturing process, which can be limiting for certain applications. 3D printing allows for the production of batteries with complex shapes and sizes that are tailored to the specific needs of the application. This could be particularly useful for the design of portable electronic devices, which often require batteries with specific shapes and sizes to fit into the available space. As shown in Figure 1A, 3D printed batteries mainly have the following four advantages, namely High Design Freedom, High Areal Energy Density, High Areal Power Density, and Flexible Manufacturing. Currently, 3D printed batteries mainly have three configurations, namely film, interdigitated, and framework, as shown in Figure 1B. The Film configuration is the simplest and has the lowest difficulty in design, fabrication, and packaging. The interdigitated configuration is mostly used in the development of solid-state batteries. The Framework configuration has the highest porosity and specific surface area, which is very helpful for improving the surface energy density and power density.

To make this paper more comprehensive and detailed, an analysis method based on big data is adopted to analyze the development and application of 3D printing in the battery field. Big data allows for the analysis of large amounts of information in a short period of time and the incorporation of multiple sources of information into one work. By leveraging the power of

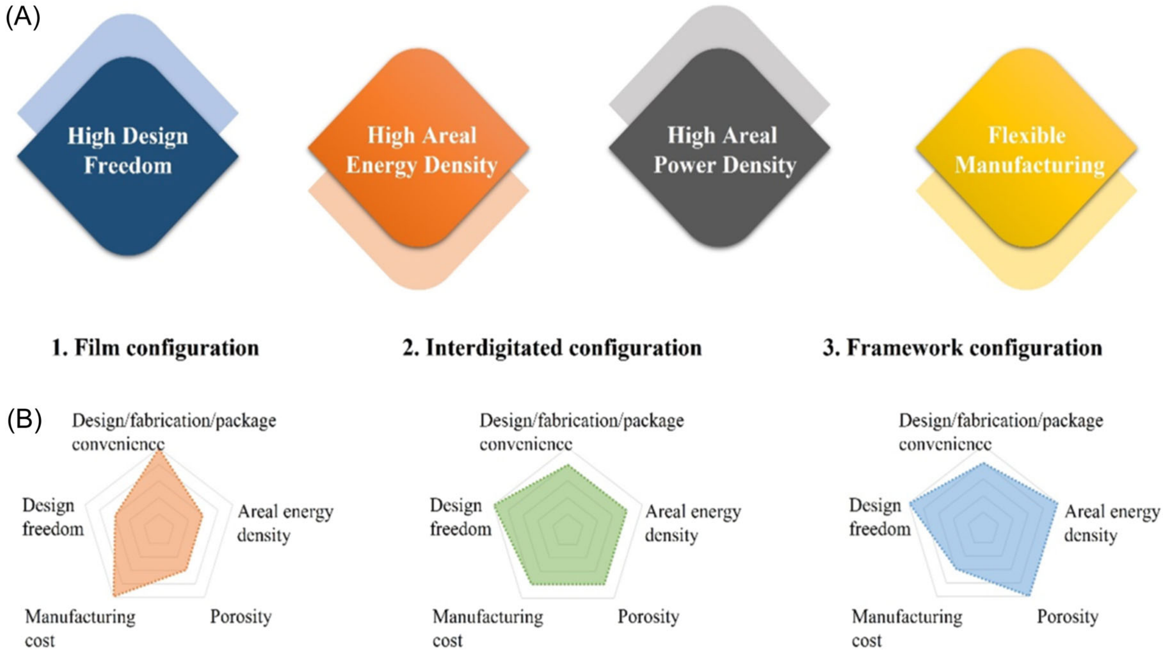

FIGURE 1 (A) A summary of advantages of 3D printed batteries. (B) Radar charts grading the performance parameters of 3D printed batteries based on different configurations. 3D, three dimensional.

big data, researchers can more easily and effectively synthesize and summarize the current state of knowledge of a particular topic, providing valuable insights and recommendations for future research. Based on big data, this review analyzes the latest breakthroughs of different 3D printing technologies, the possibility of applying different 3D printing technologies to energy storage devices, especially ZIBs electrodes and electrolytes, and some challenges faced by the existing technologies.

2 | BIBLIOMETRIC ANALYSIS

Bibliometric analysis is a rigorous and scientific computer-assisted review methodology for evaluating large amounts of data by examining key authors or authorities as identified by the majority of papers on a certain topic or field.[53] It allows researchers to investigate the subtleties of a particular field's evolutionary history while illuminating its frontiers. However, the application in battery studies is still somewhat new and underdeveloped. According to Donthu et al.,[54] the existing literature in this field is extensive and suitable for identifying evolution and literature networks. In other words, summarizing large amounts of bibliometric data to portray the intellectual structure and emerging trends is pretty much the agenda for the battery topic. The study has searched papers with the keywords "battery" and "3D printing" in the scope of the Web of Science. We then excluded nonrelevant topics in the process of data cleaning (e.g., supply chain, dentistry and oral medicine, diabetes, and healthcare policy). Finally, 445 papers that met the requirements were entered into the bibliometric analysis as original data.

2.1 | Science mapping

Prolific research analysis by country is the assessment of productive research in existing papers for various countries.[55] The top 10 countries in the data set in terms of the number of papers have been selected for further systematic analysis (see Figure 2A). The analysis presents that the number of studies published in this field in China has been increasing year by year and has overtaken the United States to take the first place for the time being. Furthermore, the total number of publications in this field in the United States and the

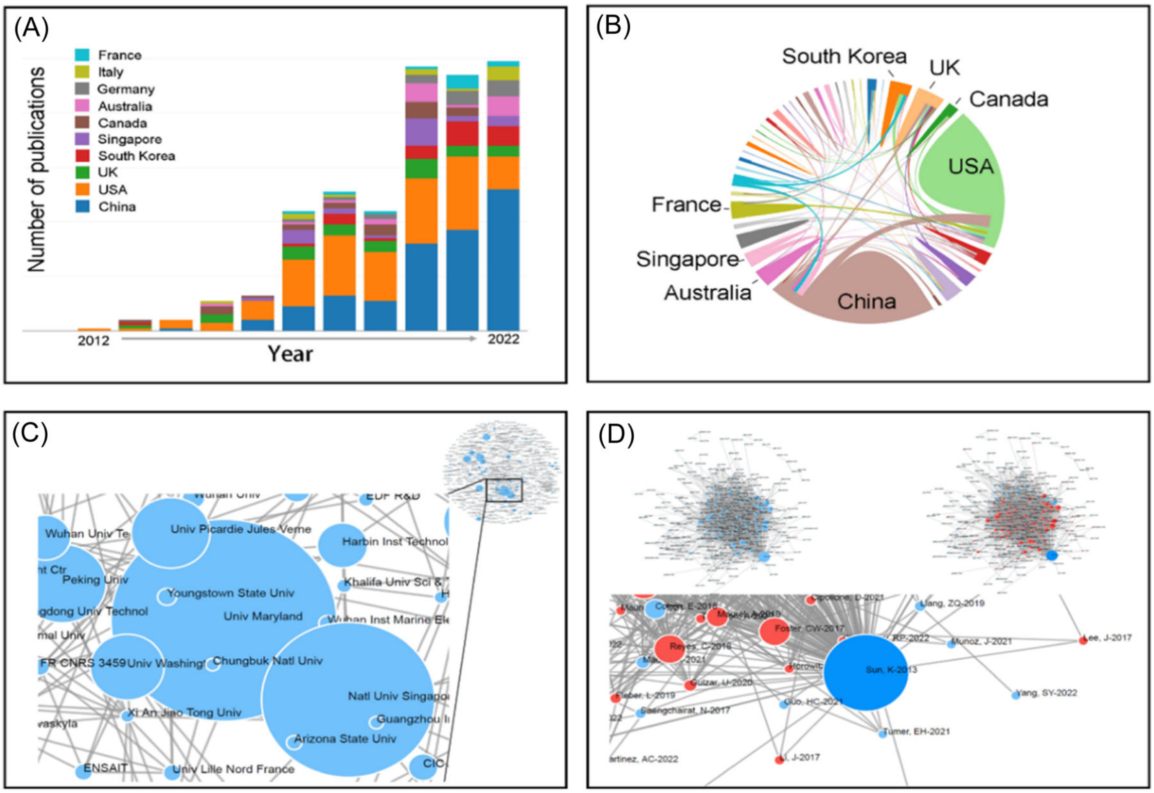

FIGURE 2 (A) The number of publications and development trends on 3D printing batteries in the top 10 countries. (B) Country collaboration network in the major countries. (C) Institution collaboration network on 3D printing battery. (D) Citation distribution of the publications of 3D printing battery. 3D, three dimensional.

United Kingdom has remained stable year on year, at around 10 and 20, respectively. Social network analysis enables a thorough examination of international collaboration, and single countries would be depicted as color blocks in a network of international collaborations (see Figure 2B).[56] A connection exists between two nations if researchers from academic institutions located in those nations contribute to at least one paper. The more coauthorship collaborations between two countries, the thicker the lines joining the color blocks in the network. The analysis reveals that China and the United States cooperate most frequently among the main countries analyzed. At the same time, the United States has also established good cooperative relations with South Korea, the United Kingdom, and Canada.

As shown in Figure 2C, institutional collaboration network analysis shows the level of collaboration between various academic institutions to which the majority of authors belong.[57] Currently, the majority of the publications about 3D printing for batteries are about LIBs. The more intense the institutional collaborations are, the thicker the edges. The study has incorporated multiple universities and research institutes in different countries into the analysis and found them intricately connected, forming small groups. The analysis unveils that the University of Maryland is the most influential institution in the field and has collaborative relations with several research institutions/universities, such as Peking University, Harbin Institute of Technology, National Institute of Aerospace, and NASA Langley Research Center. It is also worth noting that there are some universities/institutions in the lower right graph that are not as influential but work quite closely together, such as the University of Texas at Austin, the University of Technology Malaysia, Texas A&M University, and so on. According to the citation network analysis, as shown in Figure 2D, the number of citations research receives from other papers often symbolizes the prominence of

TABLE 1 Influence of institutes in the field of 3D printing batteries.

Institute	Number of papers	Total citations	Average citations	Number of first author	First author citations
University of Maryland	15	290	19.33	7	119
Harvard University	9	229	25.44	3	70
University of Illinois	7	218	31.14	5	185
National University of Singapore	13	218	16.77	3	60
State University of New York at Buffalo	6	200	33.33	4	100
University of Western Ontario	10	199	19.9	5	64
Nanyang Technological University	16	146	9.13	8	71
Korea Advanced Institute of Science and Technology	3	142	47.33	2	9
Beihang University	4	120	30	4	120
Shenzhen University	20	106	5.3	9	60
Peking University	10	101	10.1	3	49
Michigan State University	3	99	33	1	33
University of Picardy Jules Verne	16	89	5.56	4	29
University of Washington	6	82	13.67	1	0
Argonne National Laboratory	2	81	40.5	0	0
Nankai University	5	80	16	1	20
Imperial College London	5	75	15	2	29
Trinity College Dublin	8	75	9.38	1	13
Soochow University	8	74	9.25	8	74
Manchester Metropolitan University	6	70	11.67	4	59
University College Cork	6	64	10.67	3	32

Abbreviation: 3D, three dimensional.

that particular area, and other common approaches for synthesizing research into reviews (e.g., systematic reviews, meta-analyses) cannot detect these underlying metrics. Our analysis of the citation network reveals that the paper by Sun et al. dominates this field and significantly influences the development of other scholars' research.[58]

2.2 | Performance analysis

For influence analysis by institution, some significant facts are worth noting, as shown in Table 1. Shenzhen University, with a total of 20 papers, becomes the institution with the most research findings in this field. Moreover, with 290 citations, the University of Maryland is the most influential institution in this field. In addition, with 185 citations as the first unit, the University of Illinois Urbana-Champaign is the most creative research-leading institution in this field.

For influence analysis by journal, as shown in Table 2, three journals perform very strongly compared with other journals. In particular, *Advanced Functional Materials* has published 19 papers, the most supportive journal in this field. *Advanced Materials* has been cited 404 times in total, making it the most influential journal in this field.

3 | 3D PRINTING IN ENERGY STORAGE DEVICES

The manufacture of electrodes and electrolytes for energy storage devices such as batteries and supercapacitors has increasingly utilized 3D printing.[59] These 3D-printed electrodes provide a number of advantages over conventionally manufactured electrodes. One of the primary advantages of 3D printing electrodes is the ability to readily adjust the electrodes' shape and geometry to meet the requirements of the energy storage device.[60] This can enhance the device's performance and efficiency. A further advantage of 3D printing is the ability to use a variety of materials, such as conductive polymers, metals, and ceramic powders, to construct electrodes with varying qualities and capabilities.[61] This enables the creation of creative new energy storage devices with improved performance and longer lifetimes. In addition, 3D printing can dramatically cut the cost and time necessary to produce electrodes, as it enables the manufacturing of complicated shapes and structures without the need for costly molds or dies.[62]

3.1 | 3D printing technology

As mentioned in the previous section, 3D printing is the creation of actual 3D objects by successive stacks of different ink materials. The technology has its origins in printing layer by layer from computer-aided design (CAD) renderings.[63] 3D printing has come a long way in many fields such as aerospace (precision equipment or components in spacecraft), medical (artificial organs, preoperative lesion models, drug development), art (accurate printing of designer drawings), marine (model making in the design phase of ships, production of small quantities of customized ancillary products), automotive (parts of vehicles,), and so on.[64–68] The high degree of freedom, the ability to produce customized parts in small batches, and the direct control of the computer make it possible to produce almost any model designed on the computer. Nowadays, more and more materials are available for 3D printing, and different printing methods have been developed for different materials.

3.1.1 | Inkjet printing (IJP)

IJP is a low-temperature, low-pressure process used in many applications due to the wide choice of ink materials available.[69] A computer precisely controls ink nozzles to extrude the ink material and deposit it onto the substrate to form a deposited layer. In contrast, multiple layers are cured in successive deposits (the curing process varies depending on the ink material). The final product is formed once the various layers have been deposited and cured. Practically, the 3D printer can be divided into four parts according to its function: the jetting system, which is responsible for jetting the ink, the motion system, which moves the nozzles, the viewing system, and the environmental factor control system. The injection system works by using the viscosity of the liquid and the inertia of movement to eject the ink from the nozzle. The piezoelectric ceramic acts as a driver to give the nozzle an acceleration to move downwards, where the viscous force of the liquid plays a significant role; the ink moves with the nozzle and has the same acceleration; after moving to a specified position, a considerable acceleration is given to accelerate the nozzle backward; the nozzle stops moving and impacts back so that the liquid in the nozzle will be ejected from the nozzle due to the inertia of movement. The liquid in the nozzle is then removed from the nozzle due to the inactivity of movement, forming droplets that fall on the substrate at the specified location. As the injection and movement system is 3D and the controlled movement range is in the order of microns, the printed results are

TABLE 2 Influence of journals in the field of 3D printing batteries.

Journal	Number of papers	Total citations	Average citations
Advanced Functional Materials	19	194	10.21
Energy Storage Materials	13	105	8.08
ACS Applied Materials & Interfaces	13	88	6.77
Advanced Materials	12	404	33.67
Journal of Power Sources	10	67	6.7
Advanced Energy Materials	9	196	21.78
Journal of Materials Chemistry A	9	97	10.78
ACS Nano	9	63	7
Small	9	12	1.33
Nano Energy	8	109	13.63
Chemical Engineering Journal	8	25	3.13
Journal of the Electrochemical Society	8	21	2.63
Micromachines	6	1	0.17
Scientific Reports	5	65	13
ACS Applied Energy Materials	5	48	9.6
Batteries & Supercaps	5	33	6.6
Advanced Materials Technologies	5	22	4.4
Ceramics International	5	17	3.4
Applied Materials Today	5	17	3.4
Additive Manufacturing	4	32	8

Abbreviation: 3D, three dimensional.

guaranteed high quality. The most significant advantage of IJP is its broad applicability, thanks to the tolerance of ink materials and the adaptability of print resolution. Print resolution is also an essential parameter in measuring 3D printing systems, which determines the clarity and reproduction of the final product.[70] For 3D IJP technology, the size of the droplets formed by the ink material at the nozzle determines the resolution size generally; the smaller the nozzle, the smaller the ink droplets and the higher the clarity.

IJP demands research on ink engineering and binders. Wang et al. performed a series of printability tests on Zn/MnO_2 batteries using various ink types.[52] Figure 3A depicts an example pattern printed with cathode ink formulations including carboxymethyl cellulose and styrene-butadiene rubber (CMC/SBR) binders on stainless steel foils, while Figure 3B depicts a representative pattern printed with ink formulations containing modified styrene-butadiene rubber (PSBR) binders. Due to the low yield stress of the CMC/SBR cathode inks (similar to that of the PVDF-HFP cathode inks), the majority of the active ingredients, including MnO_2 and acetylene black, were readily squeezed to the edge of the printed pattern when the ink was transferred to the substrate. This resulted in the deposition of "inactive regions" in the battery electrode, which might lead to a decline in electrochemical performance. As seen in Figure 3B, outstanding printing quality with sharp edges and great uniformity was obtained using PSBR-based inks that met the defined yield stress and ink structural characteristics parameters. Figure 3C shows the flexographically printed PSBR-based MnO_2 films

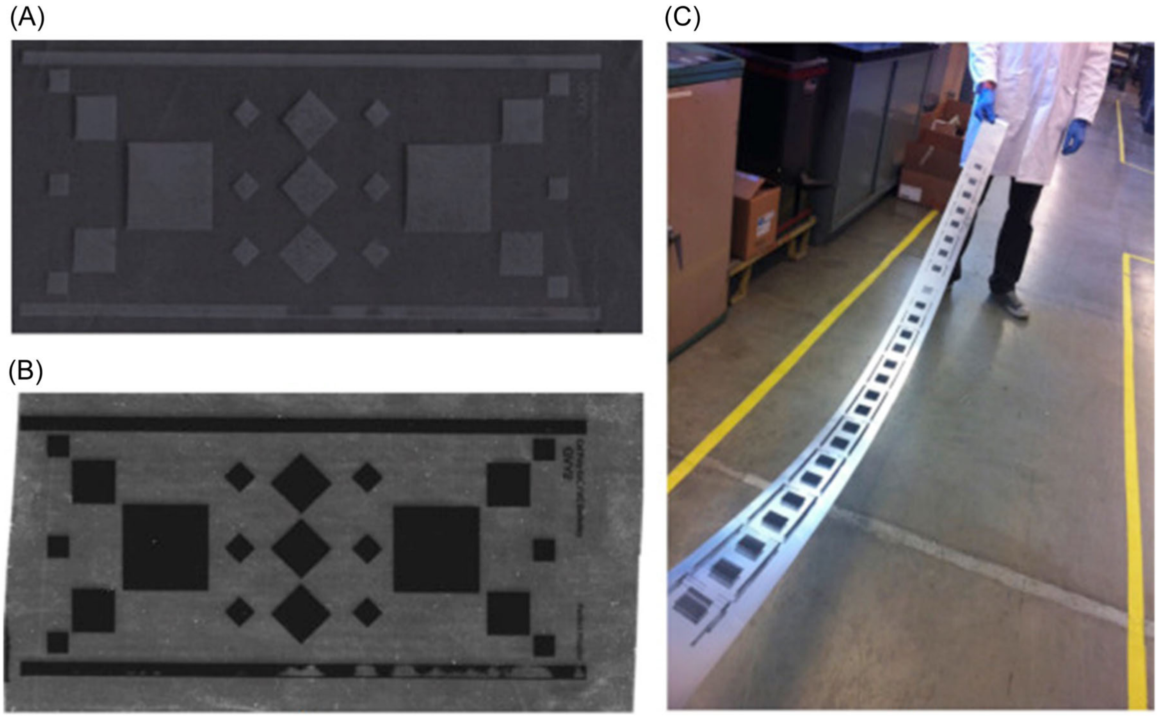

FIGURE 3 Significant enhancement in printing quality on stainless steel foils with cathode slurry inks with various polymeric binder solutions: (A) Printed films from MnO_2 cathode ink formulations with carboxymethyl cellulose and styrene-butadiene rubber binder. (B) Enhanced printed films from cathode ink formulations with PSBR binder. (C) Five continuously printed PSBR cathode films on the stainless steel foil. Reproduced with permission: Copyright 2014, Elsevier.[52]

with square patterns on a roll of stainless-steel foil after five continuous printing.

3.1.2 | Binder jet printing (BJP)

In terms of the process, BJP is an additive manufacturing method and does not require a light beam.[71] BJP allows a liquid adhesive to be sprayed directly onto a layer of powdered material, which is then joined together and densified to produce a finished product. The mainstream binder jet 3D printing technology can be divided into two types, one for the manufacture of metal parts and one for the manufacture of sand-based parts. The main process of sand-based binder jet (SBJ) printing is to distribute a thin layer of granular powder material onto a bed of powder, which is then moved using multiple, evenly distributed nozzles on top of the powder bed under precise computer control. The ink material in the nozzles is a binder that binds the powder material. The droplets of the binder are sprayed onto the designated powder area and deposited to bind the powder, and if coloration of the target material is required, an additional print head is used to spray the colorant after the liquid binder has been deposited. This is the first layer of material to be printed, and after the first layer has been printed, the powder bed is lowered to the height of this layer, and then a second layer of powder is distributed over the first layer, and the process is repeated until the part is printed. At the end of the process, the part is left in the 3D printer to allow the adhesive to fully cure, and then it is removed to clear the uncured particles from the surface, and the part is finally produced.[72] The metal binder jet (MBJ) printing process is similar to that of SBJ, except that postprocessing (infiltration or sintering) is required to reduce voids and increase the density of the prepared part to provide sufficient mechanical properties to carry the load. This means that after the part shape has been printed in the 3D printer, it needs to be transferred to a curing oven for curing, where the uncured particles are removed from the surface and after which the part is then infiltrated or sintered.[73] BJP can print on many materials including metals, ceramics, silica, sand, plastic polymers, and so on. BJP technology is very low cost in terms of hardware and materials used, and it is very fast. However, the BJP technology presents a technical challenge: the parts produced do not have excellent mechanical stability because the powder particles are bound together using a water-based binder, which leads to a loose molecular structure. Using MBJ-produced metal parts followed by sintering or infiltration can increase the density and mechanical properties of the metal part, but it will be difficult to achieve precise dimensional control.

3.1.3 | Direct ink writing (DIW)

DIW is a classical technique of extruding ink and using its rheological properties to deposit it onto a specified area of the substrate.[74] It has a wide range of applications and can be used with almost any kind of material, including ceramics, metals, polymers, nanomaterials, biomaterials, hydrogels, resins, conductive materials, and so forth. This 3D printing method is accepted in many fields because of its excellent universality and is also widely used in the design of thick electrodes and electrolytes for energy storage devices. The main process of DIW is divided into four steps, designing and building a model of the 3D object, preparing the ink with good rheological properties, printing, drying, and curing. There are two basic types: continuous IJP and droplet jetting. The most important aspect of DIW is the rheological ink, which is continuously extruded onto the substrate layer by layer, building and maintaining the framework and structure of the 3D object through the rheology of the ink rather than through drying or solidification. The ink needs to be in solid form without pressure or at low pressure, with increasing viscosity (shear thinning) as the pressure increases, so that the ink can flow smoothly through the nozzle without obstruction.[75] The inks with high viscosity and elasticity at high pressures are suitable for the DIW process because low-viscosity inks can easily form droplets when pressurized, which are difficult to form the required 3D structure on the substrate, and because inks with high viscosity at low pressures can easily clog the nozzles and require higher pressures to squeeze the inks to ensure proper DIW operation. Computer-aided design is still the main reason for the accuracy of DIW printing; the size of the nozzle diameter, the pressure applied to the ink, and the nature of the ink itself all combine to determine the resolution of the print. Layered porous frameworks and interleaved structures are not difficult for the DIW process. Such a structure would be very conducive to ions interaction and electron transport, and would also be ideal for electrolyte penetration, facilitating oxidation and reduction reactions at the electrodes. Thus, the structure would have great potential for high energy density and power efficiency. However, printing inks with good rheological properties and shear-thinning properties remain a difficulty in material selection for this technology.

3.1.4 | Fused deposition modeling (FDM)

The FDM uses thermoplastic materials such as metals, ceramics, polymers, wax, AVS, PC nylon, composite, and hybrid.[76] These materials are heated and melted in the nozzle. Melting processes include electron beam melting, selective laser sintering, selective thermal sintering, and other techniques that fuse ink materials, then move along a specific trajectory according to a digital computer model and extrude the molten material into a specified position, where it rapidly cures and adheres to the surrounding material, starting from the bottom and working in layers until it is finished. In 1987, Carl Deckard[77] developed the selective laser sintering technique, which is fast and accurate and can work with metals, plastic polymers, and ceramics. This technique is also part of the 3D printing technology, where thermoplastic powders are heated and melted to create 3D printed objects. Polylactic acid (PLA) is the most widely used material in FDM technology and is inherently nontoxic, has a relatively low melting point (around 176°), and is also very compatible with living organisms. For FDM, there are now two aspects, one is the technical research on the optimization of process parameters to improve the bonding properties of the material interface as much as possible and to improve the mechanical properties; the other direction is the research on the modification of PLA to enhance the degree of crystallization of PLA molecular chains, improve their interaction state, and enhance the mechanical properties of PLA. The increased functionality of PLA can bring features such as thermal stability, good electrical conductivity, and antistatic properties to components containing PLA, which can greatly enrich the applications of PLA components.

3.1.5 | Directed energy deposition (DED)

Unlike the material extrusion type of 3D printing, DED uses materials in the form of powders or wires, which are melted by laser or ion arc, or electron beam techniques as the extruded material is deposited on the substrate and then printed and cured layer by layer.[78] Coaxial and lateral feeding technology is crucial for DED technology, as the core of DED technology is focused on thermal energy, so the direction of material transport and deposition conditions are very important for DEDs with axial feeding devices.

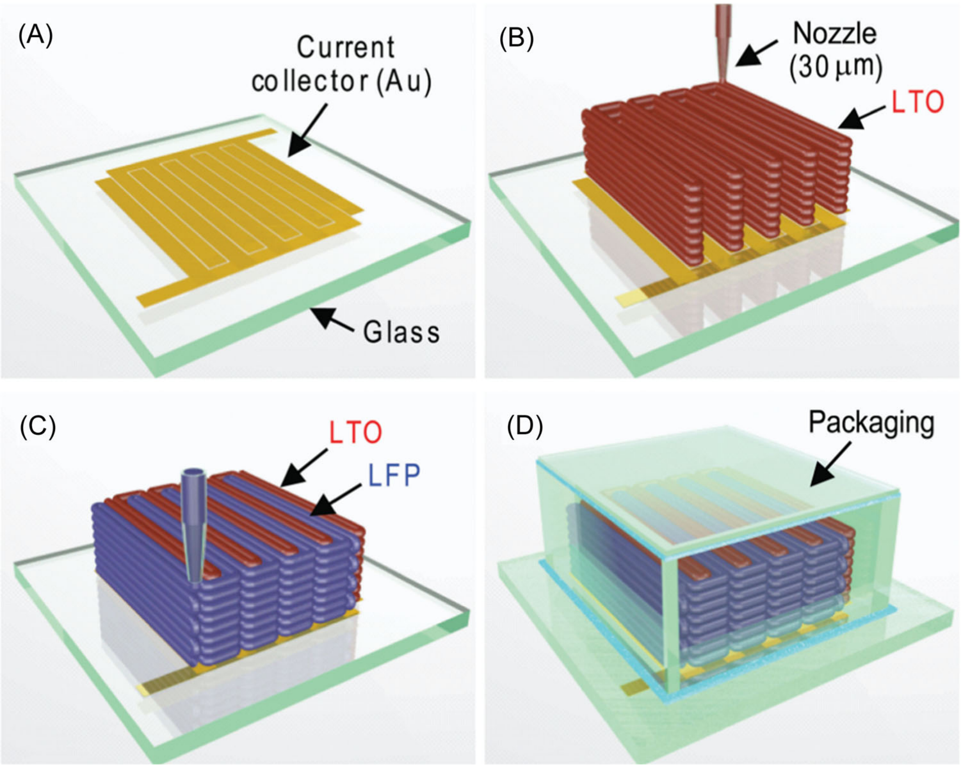

FIGURE 4 Schematic diagram of the 3D printing process. (A) Printing on the gold collector (B) LTO and (C) LFP by 30-micron nozzle, followed by sintering and (D) finally packaging.[82] Reproduced with permission: Copyright 2013, John Wiley and Sons.[82] 3D, three dimensional; LFP, $LiFePO_4$; LTO, $Li_4Ti_5O_{12}$.

3.1.6 | Other 3D printing technologies

Stereolithography (SLA)

SLA equipment usually uses materials that are liquid resins or other photopolymers that will cure under UV light.[79] Photo-curing resins or other photopolymers can be converted from liquid to solid by a chemical reaction using light with wavelengths in the visible or ultraviolet range. After the material has completed its transformation from liquid to solid, a layer of material is formed and stacked on the substrate. By repeating the above actions repeatedly, layer upon layer of material is stacked to form the shape of the part. After printing, cleaning is required to remove any uncured or incompletely cured resin material. SLA equipment is very accurate, being somewhat more precise than other 3D printing methods, but it also has many drawbacks. SLA can only print on materials that are light and curable, and these materials usually have limited conductivity, so most conductive polymers cannot be used directly in SLA. The electrochemical properties are essential to produce electrodes, and the lack of electrochemical properties of light-curing materials (photo-curing resins) or photopolymers requires the doping of other materials to be able to function as electrodes. This limits the use of SLA in terms of production costs and handling difficulties, as well as the possibility of producing toxic substances that are harmful to the environment during the printing process.

Spin-coating

Spin coating is a ubiquitous 3D printing method for fabricating flexible batteries.[80] The configured solution is spin-coated onto the substrate, and other components are added layer by layer, followed by annealing. This is a very efficient method. The spin-coating method can also be applied to the production of medical models of different sizes, complexity, and prescribed thickness, which will help physicians develop surgical treatment strategies based on the prepared medical models. Ching-Zhuo Chi and Li-Zhong Mu, et al. used silicone to produce patient-specific vascular models by continuous coating and the accurate models of abdominal aneurysms were produced with an error of less than 2 mm compared with measurements from computed tomography images.[81] However, since the spin-coating method cannot accurately control the amount of each material, the prepared films usually have an uneven thickness, uneven surface, low efficiency and instability, and short life. Another drawback is that the materials may interact with each other. Therefore, the spin-coating 3D printing method is only suitable for certain materials or special cases.

Material injection

In material jetting, droplets of material are deposited from the print head onto the substrate using thermal or piezoelectric methods, after which the deposited droplets solidify and are dropped into a second layer; each layer is built on top of the previous one, which is then cured by UV light. This 3D printing technology can make smooth, precise parts, and polymers, ceramics, composites, and even biomaterials are among the materials that can be applied in material jetting technology.

3.2 | 3D printed energy storage devices

Based on the literature analysis, the 2013 publication on Li-ion micro battery architectures is the most concentrated paper in the 3D printing battery field. Sun et al. described a kind of LIBs that were fabricated via 3D printing, as shown in Figure 4A–D. Initially, Sun et al.[82] used $Li_4Ti_5O_{12}$ (LTO) as the anode of the battery and $LiFePO_4$ (LFP) as the cathode of the battery to manufacture the LIBs using a 3D interleaved microcell architecture. This paper uses a graded volatile solvent system owing to the variable solidification durations of various solvent materials after 3D printing and to make different solvents perform diverse roles. Maintaining a temperature of 100°C during the printing process evaporates the water, causing the printed part to lose its water solidification and ensuring its integrity. The boiling point of ethylene glycol is at 197.3°C, while the boiling point of glycerol is at 290°C, neither of which will evaporate during the printing process. Therefore, ethylene, glycol, and glycerol are used as humectants to ensure good adhesion.

In recent years, 3D printing has extended to a wider field. Zong et al.[83] proposed for the first time in 2021 a topochemistry-driven method for constructing expanded 2D rhenium selenide intercalated by nitrogen-doped carbon hybrid (E-$ReSe_2$@INC) with a strong-coupled interface and weak van der Waals forces to achieve a high energy/power density 3D-printed sodium-ion hybrid capacitor (SIHC). As a new additive manufacturing process, 3D printing technology based on extrusion may be utilized to produce SIHCs devices with bespoke architectures (Figure 5A). To ensure a successful 3D printing process of electrodes for high-performance SIHCs devices (3DP-SIHCs), the rheological properties of E-$ReSe_2$@INC/carbon nanotube (CNT)/graphene oxide (GO) ink and active carbon (AC)/CNT/GO ink were assessed. As seen in Figure 5B, both E-$ReSe_2$@INC/CNT/GO and AC/CNT/GO inks exhibit non-Newtonian fluid-like apparent viscosities and shear-thinning tendencies, allowing for continuous flow of printable inks during extrusion processing. Excellent rheological qualities enable the E-$ReSe_2$@INC/CNT/GO, and AC/CNT/GO inks may be extruded smoothly under shear stress to create a steady flow of ink during the extrusion printing process without clogging. Figure 5E shows the cyclic voltammetry (CV) curves of the E-ReS_2@INC/AC 3DP-SIHCs device at different scan rates. Even at a high scan rate of 100 mV/s, the device retains a nice shape. The E-ReS_2@INC/AC 3DP-SIHCs achieve an areal energy and power density of 0.32 and 38.76 mW/cm^2 and a volumetric energy density of 3.2 mWh/cm^3, which are superior to the majority of the capacitor and battery systems manufactured by different printing processes (Figure 5C). In light of the strong temperature resistance properties of E-ReS_2@INC in Na-ion storage, the 3D-printed E-ReS_2@INC/AC SIHCs device likewise demonstrates great performance throughout a broad temperature range (Figure 5D). Based on this method, Zong et al.[84] carried out extended work to effectively improve the rate performance and stability of potassium-ion hybrid capacitors through size control, as shown in Figure 5E. The heavily exposed three-phase boundary (MoP-C-electrolyte) facilitates the transport and storage of K ions. This strategy can significantly enhance the energy density and cycle stability of energy storage devices, which is meaningful for the material development and preparation methods of next-generation energy storage devices.

4 | 3D PRINTING STRATEGY IN ZIBS

Zn is an abundant and widely available element, making ZIBs more environmentally friendly than other types of batteries that rely on rare or toxic materials.[85] ZIBs have a number of advantages over traditional LIBs, including lower cost, high energy density, and high safety. 3D technology has been increasingly used in the development of ZIBs, due to its ability to easily customize the shape and geometry of the electrodes and allow for the use of a wide range of materials.[86] 3D printing has been used to produce complex and high-performance electrode structures, such as hierarchical and sponge-like architectures, which can improve the electrochemical properties of ZIBs.[87] Electrodes are a vital component in cyclically rechargeable batteries. The development of electrode materials for ZIBs has made significant strides in recent years.[88] In addition to the material of the electrode, the configuration design has an effect on the

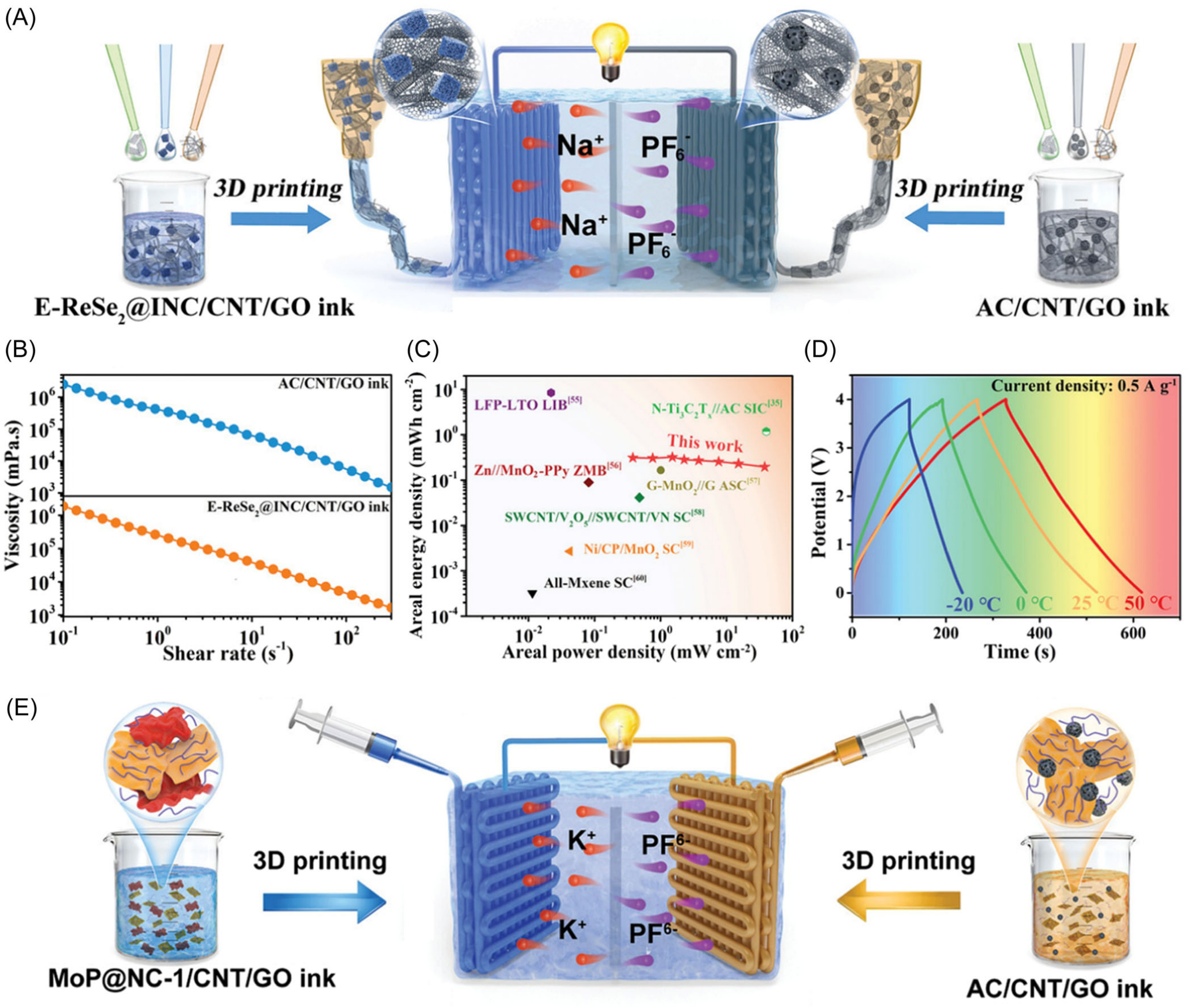

FIGURE 5 Fabrication of E-ReSe$_2$@INC//AC 3DP-SIHCs. (A) Schematic illustration for E-ReSe$_2$@INC//AC 3DP-SIHCs device and corresponding charge process. (B) The apparent viscosity as a function of shear rate. (C) Ragone plots of the E-ReSe$_2$@INC//AC 3DP-SIHCs in comparison with other reported energy storage devices. (D) Galvanostatic charge-discharge curves tested at 0.5 A/g of 3DP-SIHCs device at various temperatures. Reproduced with permission: Copyright 2022, John Wiley and Sons.[83] (E) Schematic illustration for MoP@NC-1//AC 3DP-PIHCs device and corresponding charging process. Reproduced with permission: Copyright 2021, John Wiley and Sons.[84] 3D, three dimensional; AC, active carbon; PIHC, potassium-ion hybrid capacitor; SIHC, sodium-ion hybrid capacitor.

power of the ZIBs.[89–91] A superior configuration may substantially enhance the utilization of the electrode material and maximize the oxidation or reduction reactions.[92] 3D printing technology is well-suited for the creation of electrodes for ZIBs since there are currently several advanced 3D printing technologies for a variety of materials and ink engineering technologies that can preprocess the target material. The precursors of the target material can be fabricated to enable the 3D printing technology to be adapted to the electrode material of the ZIB. Due to the diversity of 3D printing methods, the conditions involved (such as high temperature, and strong light) need specific requirements for the precursor liquid. And the uniform distribution of materials (binders, electrode materials, electrolytes, etc.) affects the performance and stability of 3D printed ZIBs. Moreover, the 3D printing technology can be precisely guided by CAD, which can accurately create 3D models of complex ZIBs electrodes and increase the efficiency of the 3D printed ZIB electrodes.

There are many new 3D printing technologies that have been applied to the manufacture of ZIBs, such as IJP, FDM, DED, DIW, and so forth, all of which can be utilized in the manufacture.[93–95] In recent years, ZIBs

TABLE 3 Printed ZIBs based on different cathode materials and the electrochemical performance of electrodes.

Cathode material	Specific capacity (mAh/g)	Average working voltage (V vs. Zn^{2+}/Zn)	Energy density (Wh/kg)	Printing method	Reference
Manganese oxide					
α-MnO_2	260	1.25	350	DIW	[97]
β-MnO_2	320	1.38	400	Stencil printing/DIW	[98]
ε-MnO_2	280	1.26	385	Stencil printing/DIW	[98]
δ-MnO_2	270	1.22	330	Stencil printing/DIW	[98]
γ-MnO_2	290	1.23	380	DIW	[99]
$Zn_xMnO_2 \cdot nH_2O$	300	1.28	390	DIW	[100]
PANI-MnO_2	275	1.41	375	IJP	[101]
Mn_3O_4	250	1.45	315	DIW	[98]
Vanadium oxide					
VO_2	260	0.72	275	Spin coating	[102]
V_2O_5	475	0.75	330	Electrospinning	[102]
$V_5O_{12} \cdot 6H_2O$	380	0.74	310	DIW	[103]
$Ni_{0.25}V_2O_5 \cdot H_2O$	400	0.77	290	DIW	[104]
$Li_xV_2O_5 \cdot nH_2O$	410	0.52	280	DIW	[104]
$NaV_3O_8 \cdot 1.5H_2O$	390	0.65	270	DIW	[104]
$K_2V_6O_{16} \cdot 2.7H_2O$	290	0.68	230	DIW	[104]
Prussian blue					
$KCuFe(CN)_6$	80	1.65	90	DIW	[105]
$Zn_3[Fe\ (CN)_6]_2$	120	1.68	110	DIW/FDM/IJP	[106]
$FeFe(CN)_6$	140	1.32	170	FDM	[106]
$CoFe(CN)_6$	180	1.66	300	FDM	[107]
Spinel-structured oxide					
$ZnAl_xCo_2$-xO_4	130	1.70	200	DIW	[108]
$ZnNi_{0.5}Mn_{0.5}CoO_4$	180	1.65	310	DIW	[108]
$ZnMn_2O_4$	160	1.31	205	FDM	[101]
Mo_6S_8	140	0.40	60	IJP	[109]
$ZnMo_6S_8$	140	0.41	60	IJP	[109]

Abbreviations: DIW, direct ink writing; FDM, fused deposition modeling; IJP, inkjet printing; ZIB, zinc-ion battery.

based on 3D printing technology have made great progress.[96] The performance of ZIBs based on different cathode materials and the corresponding 3D printing methods are shown in Table 3.

4.1 | 3D printed film configuration in ZIBs

As the simplest configuration, the design, preparation, and packaging of the film configuration are the least difficult, which gives a high degree of flexibility.

4.1.1 | Thin film configuration

Thin film electrodes have a good energy density, efficiency, and flexibility, can be bent at specific angles, and are inherently thin film in nature, making an excellent adaptation to the requirements of miniaturized batteries thereby providing a high level of universality.[110–112] As thin film electrodes can increase the reaction area of the electrode to a large extent, the actual working reaction ratio of the electrode material (the ratio of the reacting electrode volume to the overall electrode volume) can be increased significantly, thus increasing the efficiency of the electrode. Zn/MnO_2 is a very typical, printable thin film ZIBs. Mn-based oxides have good multiplicative properties and can have relatively good power densities, but the Jahn–Teller effect of the Zn ions during the embedded phase transition causes the Mn ions to dissolve during cycling, which has an impact on the lifetime of the Mn-based oxide electrode.[113] Studies now show that the addition of additives containing Mn ions to the electrolyte can inhibit the dissolution of MnO_2 electrodes, so Zn/MnO_2 is very widely used in printed batteries.[114] As a thin film electrode is used, Zn/MnO_2 type ZIBs can be fabricated using a printing method. Current technology prefers to use 2D printing methods such as screen printing, as shown in Figure 6A–D, where graphene is printed on a blank Polyethylene terephthalate substrate using graphene ink to collect the current, followed by MnO_2 ink to print the cathode and finally the anode, Zn ink.[115] These thin-film electrode-based ZIBs are now predominantly printed in 2D, as shown in Figure 6F–K; 3D printing will allow the printed electrodes to be 3D in shape, allowing for better ion interaction. Thin film electrodes are an important contributor to the development of ZIBs toward flexibility, bendability, and high efficiency, but their 2D structure (thin film) limits the ability of thin film electrodes to achieve energy and power densities in a limited space.

4.1.2 | Thick configuration

Thin film electrodes limit their own energy density because they are inherently thin film in nature, and it is

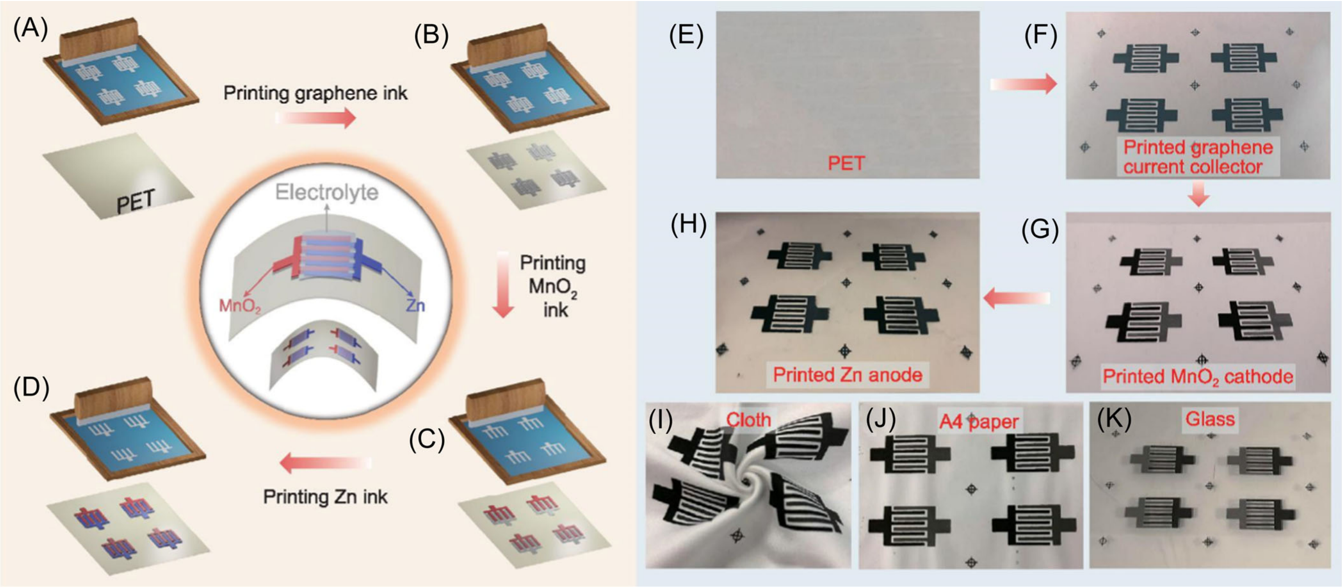

FIGURE 6 Fabrication of Zn/MnO_2 planar micro batteries by printing. (A–D) Schematic of screen-printing Zn/MnO_2 micro batteries fabrication: (A) the blank PET substrate, (B) the printed graphene current collectors, (C) the printed MnO_2 cathode, and (D) the printed Zn anode. (E) The blank PET substrate, (F) the graphene current collectors, (G) the printed MnO_2 cathode, and (H) the printed Zn anode on interdigitated graphene fingers. Zn/MnO_2 MBs imprinted on several substrates, including (I) fabric, (J) A4 paper, and (K) glass.[115] Reproduced with permission: Copyright 2020, Oxford University Press.[115] PET, Polyethylene terephthalate.

difficult to increase the volume of the electrode material and therefore the efficiency by increasing the thickness. The advent of 3D printing has ameliorated this situation. 3D printing gives the ZIBs a high degree of freedom, and for the ZIBs electrode, the increased height dimension allows the electrode to be 3D, and according to the following equation:

$$C = \frac{\alpha D}{L^2}, \tag{1}$$

where C is the rate capability, D is the diffusivity of the material, and L is the diffusion length, which is related to the spatial dimension of the material and can increase the energy density and power density of the ZIB. However, increasing the thickness of the electrode increases the distance between the Zn ion and the electron, so from the point of view of electron transport kinetics, a balance between the conductivity of the Zn ion and the electron and the thickness of the electrode needs to be achieved, for good stability and charging and discharging efficiency.[116] As the thickness of the electrode can be increased, more electrode material or other additives are required, which can be modified by adjusting the ratio of electrode material and additives to increase the electron conductivity of the electrode.

Regarding the effective contact of the electrodes, this depends to a large extent on the microstructure of the two substances. In other words, it is the effective contact between the electrodes and the electrolyte, the electronic conductor, and so forth, that affects the energy density and power density of the ZIBs, and the size of the effective contact area depends on the shape and distribution of the individual components of the ZIBs.[117] In general, electrolytes and electrodes can be understood as irregular solid particles, and solid particles are usually rigid in nature, so contact between particles can become very difficult. The Hertz contact theory enables a simple description of the contact between two elastic spheres under a force F, as shown in the following equation:[118]

$$r_0 = \left(\frac{R_1 R_2}{R_1 + R_2}\right)^{\frac{1}{3}} \frac{3}{4} F \left(\frac{1 - \nu_1^2}{E_1} + \frac{1 - \nu_2^2}{E_2}\right). \tag{2}$$

R_1 and R_2 are the radii of the two particles, E_1 and E_2 are the moduli of elasticity, and υ_1 and υ_2 are the Poisson's ratios. According to this equation, the radius, modulus of elasticity, and Poisson's ratio of the two particles can be reduced and the contact area between the particles can be increased by increasing the squeezing pressure. In ZIBs, this means that the effective contact area at the interface between electrolyte and electrode can be increased. Ideally, the electrode material should be large enough to achieve a high energy density and the electrolyte and electrode material should be in maximum contact to maximize the power density of the ZIBs. While considering the contact area between the electrode and the electrolyte, the ionic conductivity of the electrode and the negative space filled by the electrolyte also influence the mobility and conductivity of Zn ions and electrons. According to the effective ionic conductivity D_{eff} formula (Equation 3), the porosity of the electrode and the curvature of the electrode will have an impression on the mobility of the ions.[119]

$$D_{\text{eff}} = \frac{\varepsilon}{\tau_D} D. \tag{3}$$

In D_{eff} formula, ε is the porosity, τ is the tortuosity, and D is the intrinsic ion conductivity. So according to the equation, the mobility of Zn ions can be increased by increasing the porosity of the electrode and reducing the curvature. Increasing the porosity of the electrodes is a very demanding process and the thick electrodes have a "thickness" which, unlike thin film electrodes, makes it impossible to use conventional 2D printing methods. 3D printing facilitates thick electrodes with higher porosity and less curvature and now exists for the initial application of printing ZIBs electrodes.[120–122] Modern 3D printing technology is advancing at a rapid pace, and technological changes and updates undoubtedly mean that there are more materials to choose from.

4.2 | 3D printed interdigitated configuration

Interdigitated electrodes, which yield closer contact due to their characteristic alternating cathode and anode electrodes on the submillimeter scale, are considered to be suitable for increasing the surface contact in ZIBs.[123] In 2022, CNT is coated with MnO_2 using 3D printing technology to create a flexible CNT@MnO_2 ink as a cathode for flexible aqueous micro-ZIBs.[124] This work demonstrates the potential of 3D printing technology for the creation of manganese-based flexible ZIBs. Figure 7A depicts the multinozzle printer of the 3D printing system. The illustration depicts several configurations of contemporary silver collectors, including single-cell configurations, parallel configurations, and two-cell series arrangements. In the following printing phases, the shape of the battery may be selected at the discretion of the designer. As is well knowledge, the electrode material is the most important component of this sort of battery, and several parameters must be taken into account throughout the production process. In this system, the cathode material

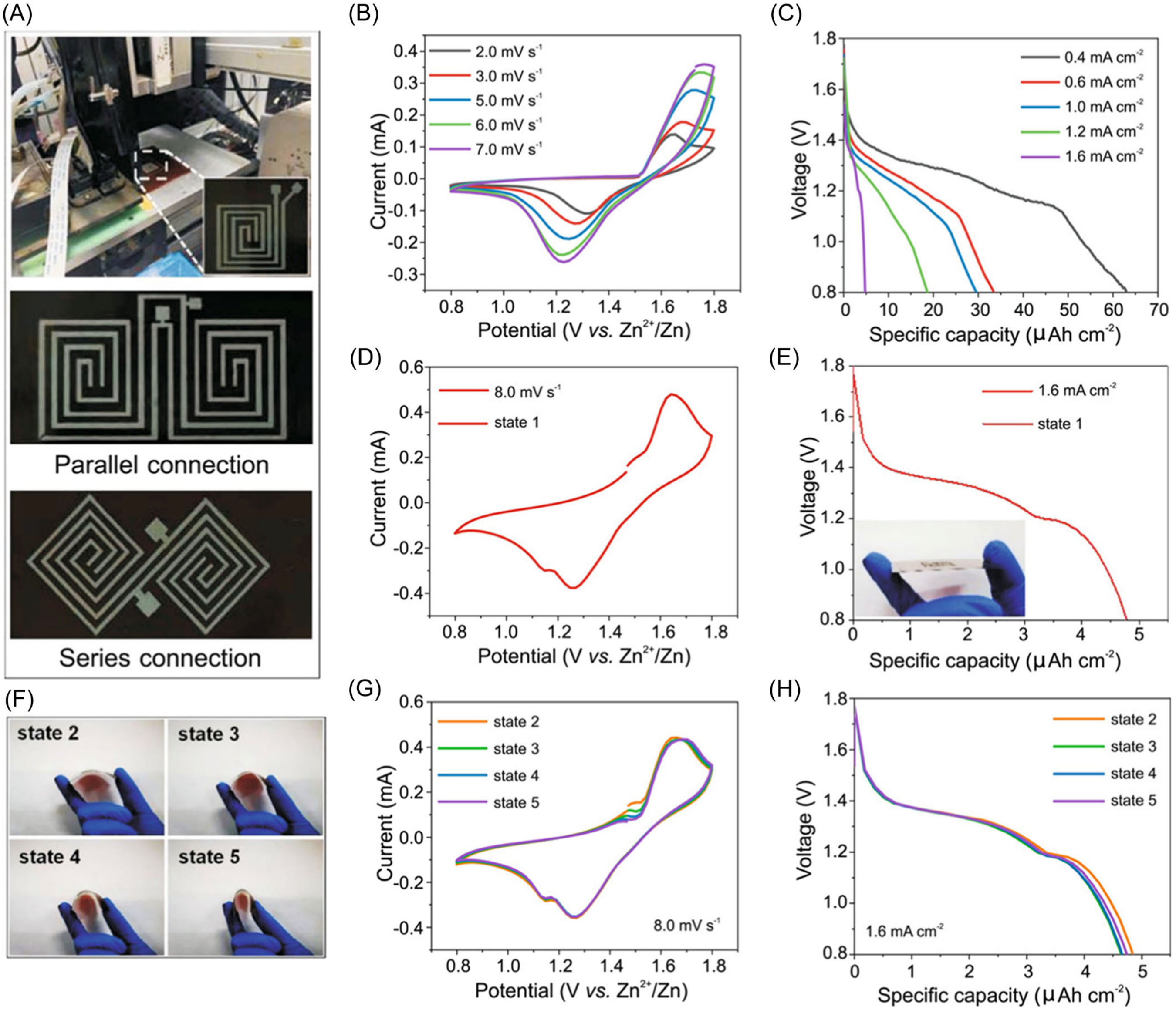

FIGURE 7 (A) Photograph of the multinozzle printing system and a heating plate of the 3D printing equipment printer; the insets show the looped current collector with a single structure, a parallel structure, and a series structure, respectively. Electrochemical properties of the 3D printing flexible batteries: (B) CV curves at different scan rates and (C) discharge profiles at different current densities. (D) CV curves at a scan rate of 8.0 mV/s in the static state. (E) Discharge profiles at a current density of 1.6 mA/cm^2 (the inset shows the photograph in the static state). (F) Photographs of the flexible battery in different bending states. (G) CV curves at a scan rate of 8.0 mV/s of the 3D printing flexible batteries under different bending states. (H) Discharge profiles at a current density of 1.6 mA/cm^2 under different states. Reproduced with permission: Copyright 2022, John Wiley and Sons.[124] 3D, three dimensional; CV, cyclic voltammetry.

is a low-hardness and low-density metal oxide. Therefore, typical grinding techniques may be used to make inks for 3D printing. For the anode, Zn powder is easily oxidized because of its high hardness, and it is processed using a ball mill. Zn powder ink is made using the typical ratio of active material to the conductive agent and the binder is prone to agglomeration. To turn ink into electrodes, an automated control system is necessary for pattern creation and the confirmation of print orders. Anode and cathode materials for printing will be delivered to the striker piezo nozzle and then dried using a heating plate with a constant temperature and negative pressure. Due to the low electrochemical stability and poor interfacial qualities of polymer electrolytes, aqueous Zn electrolytes continue to be used for testing flexible batteries. Figure 7B,C depicts the CV curves at scan rates of 2.0, 3.0, 5.0, 6.0, and 7.0 mV/s and the discharge curves at 0.4, 0.6, 1.0, 1.2, and 1.6 mA/cm^2 when the cell is at rest. Due to the vast difference in the active material loading between the 3D printed electrode and the conventional electrode, after low current preactivation, there is only a single oxidation peak. Moreover, with a current density of 0.4 mA/cm^2, the battery's discharge-specific capacity reaches 63 Ah/cm^2. To further illustrate the adaptability of the battery, the CV and discharge curves were studied in various bending states. Under static circumstances, the CV curve (Figure 7D) at a scan rate of 8.0 mV/s and the discharge curve (Figure 7E) at a current density of 1.6 mV/s^2 are shown. The specific discharge capacity of State 1 is 4.78 Ah/cm^2. Figure 7F shows photographs of the battery in various bending states, whereas Figure 7G,H depict the electrochemical curves corresponding to each condition. In general, the contour of the curves in each state is well-maintained, the curves are smooth, and there are very few peaks and valleys. States 2 through 5 have specific discharge capacities of 4.85, 4.65, 4.66, and 4.74 Ah/cm^2, respectively. The highest change of all bent states relative to the original condition is just 2.72%, confirming the battery's exceptional flexibility and strong stability. The slight variation in capacity is a result of the altered contact area between the electrode and electrolyte in the bent condition.

4.3 | 3D printed framework configuration in ZIBs

The frame configuration can provide electrodes with high porosity and specific surface area, and some advantages of this configuration are extended when applied to ZIBs.[125] Poor cycle stability resulting from dendritic formation and undesired side reactions of the Zn anode impedes the extensive commercial implementation of ZIBs. Using a combination of 3D printing and electroless plating/electroplating processes, Duan et al.[126] presented a new 3D Zn metal anode with a multichannel lattice structure. The 3D Ni-Zn anode with a multichannel lattice structure and superhydrophilic surface may effectively enhance the electric field distribution and produce uniform Zn deposition without the formation of Zn dendrites. Due to the low Zn nucleation overpotential and consistent local electric field distribution, the 3D Ni-Zn cell demonstrates highly reversible Zn plating/stripping and good Coulombic efficiency. Full cells made with 3D printed Zn anodes and polyaniline-intercalated vanadium oxide cathodes demonstrate exceptional performance. It is evident that the deposited Zn forms irregular dendrite-like particles on the surface of the 2D Ni electrode even at a modest capacity of 2 mAh/cm^2 (Figure 8A). Initial dendrite-like particles may lead to a constant charge accumulation, eventually driving the formation of dendrites (Figure 8B). When the capacity reaches 10 mAh/cm^2, a substantial quantity of Zn is continually plated in the nucleated region (Figure 8C), even generating dendrites that may penetrate the separator. In contrast, after 2, 5, or even 10 mAh/cm^2 of Zn deposition, the 3D Ni electrode's surface is still smooth and flat. Figure 8H depicts the development of the morphology of a Ni 3D electrode during Zn deposition. These findings visibly demonstrate that the electrode with a 3D multichannel lattice structure inhibits the formation of Zn dendrites on the exterior surface of the 3D Ni electrode. The simple and inexpensive production of conducting metal lattices with programmable 3D multichannel layouts paves the way for the development of batteries with additional high-performance metals (e.g., Li, Na, K, Mg, Al).

5 | CONCLUSION AND OUTLOOK

The 3D printing of improved batteries has garnered significant interest as a disruptive technology that may affect the design and architecture of materials, modules, and devices on each level. The purpose of this review is to provide readers with a complete grasp of the enormous application possibilities of 3D-printed batteries. Rapid development over the past few years has resulted in a number of technological advancements, including the optimization of the most suitable printing technology for desirable 3D battery products, as well as the modification of the battery modules, full battery level architecture, and configurations to achieve printable materials, electrodes, and electrolytes. As these technologies progress, a comprehensive knowledge of the governing principles regulating 3D printing materials, components, and printing procedures, as well as the consequent battery performance, has been sought. While considering the development of ZIBs, 3D printing brings some very considerable benefits in this field. One of the key benefits of using 3D printing technology to produce ZIBs is the ability to customize the shape and size of the batteries to fit specific needs. This could be particularly useful in the development of wearable devices or other electronic devices that require compact, lightweight power sources. 3D printing also allows for the creation of complex internal structures within the battery, such as electrodes with specific shapes or patterns, which could potentially improve the performance and efficiency of the battery. Another potential advantage of using 3D printing for ZIBs is the ability to produce them quickly and efficiently. Traditional battery manufacturing processes can be slow and labor-intensive, but 3D printing allows for the production of batteries in a matter of hours or even minutes. This could potentially reduce the cost and

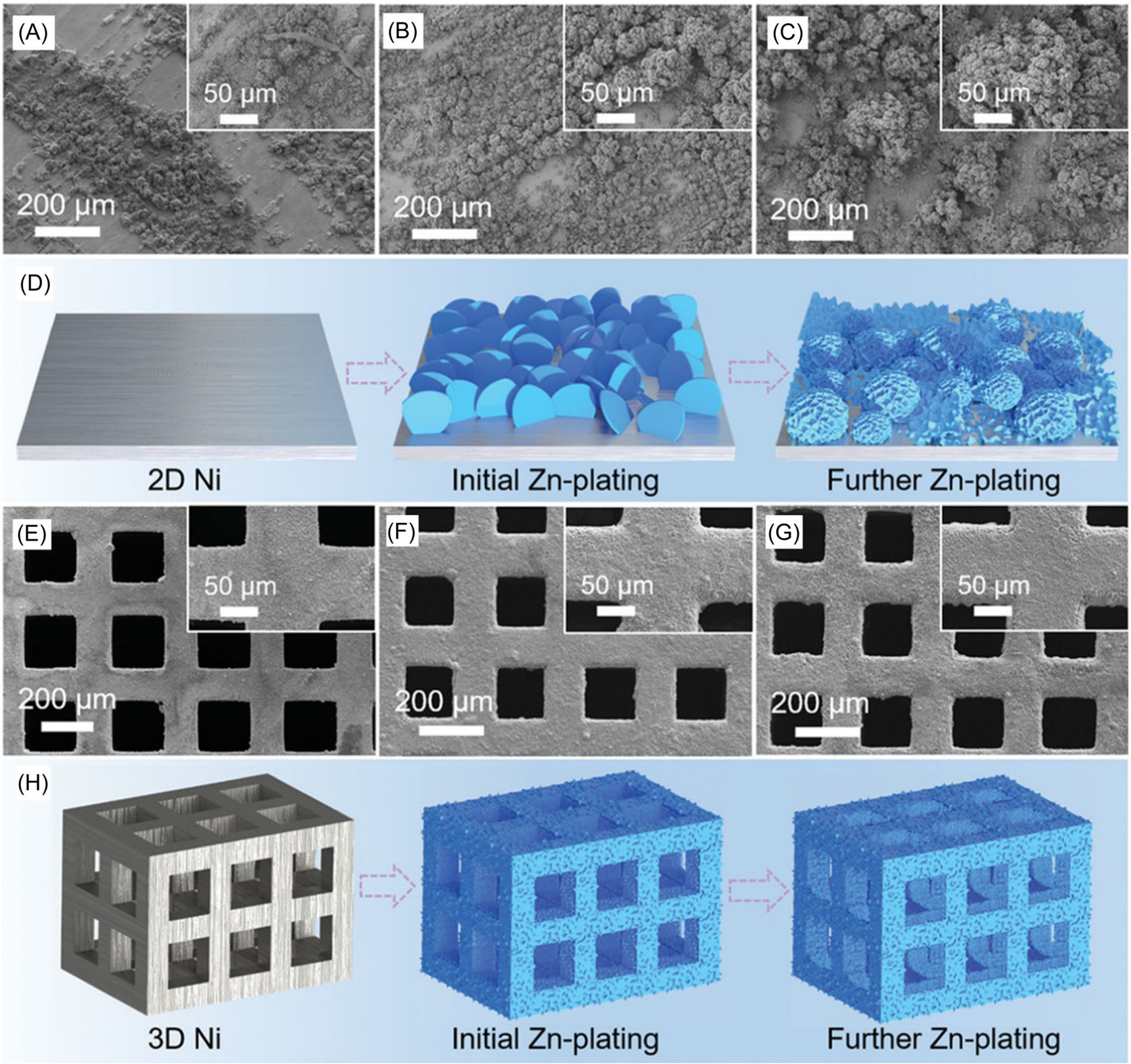

FIGURE 8 Scanning electron microscope images of Zn plating with the capacity of (A) 2, (B) 5, and (C) 10 mAh/cm^2 on 2D Ni, and (E) 2, (F) 5, and (G) 10 mAh/cm^2 on 3D Ni. The corresponding schematic illustration of Zn deposition on the (D) 2D Ni and (H) 3D Ni.[126] Reproduced with permission: Copyright 2021, John Wiley and Sons.[126]

time required to bring new battery designs to market, enabling faster development and deployment of advanced battery technologies. However, there are also some challenges and limitations to using 3D printing for ZIBs. One of the main challenges is the need to develop new materials and techniques that are suitable for use in 3D printing. Currently, most 3D printing processes are limited because of the use of polymer binders or additives, which may not be suitable for use in high-performance batteries. Additionally, there are concerns about the potential for defects or imperfections in 3D printed batteries, which could negatively impact their performance and lifespan.

For the development path and forecast for 3D-printed ZIBs production, some suggestions are listed here. First, in terms of printable materials, LIBs are the most successful printable battery material to date. The expansion of this technology to an additional energy storage device, such as ZIBs, is one of the keys to current progress. For 3D-printed batteries to reach better energy densities, active materials that are compatible with the 3D printing method must be developed; this is essential for commercial-scale applications. Future printed batteries' electrochemical performance will be enhanced by printing methods based on additive-free and binder-free electrodes. Second, the most prevalent printing process for ZIBs is DIW, and the printing resolution is often just tens or hundreds of microns. To make printed batteries more compact with finer feature sizes, it is important to create high-resolution printing processes with feature sizes as small as 1 mm. Although the high resolution is achievable with DIW printing with microcapillary nozzles, manufacturing suitable printable inks with the necessary yield stress behavior and active ingredients is a must, since it must not clog the tiny nozzles. Other developing 3D printing technologies than DIW should be studied further for battery module production. Third, in terms of printing design, a more logical design in terms of the electrode structure and battery configuration, as well as customized design and manufacturing, is necessary to further enhance battery performance. Taking into account the compatibility of ink materials and 3D printing technology, the appropriate material, 3D printing technology, and manufacturing process should be chosen in the production of ZIBs, to adapt to various environmental and technical requirements. Fourth, the performance and stability of 3D-printed ZIBs need to be optimized in terms of conditions and costs. Although some existing 3D printing and ink preparation technologies are able to produce ZIBs with relatively good energy density and electrochemical efficiency, their harsh production conditions, cumbersome preparation processes, and high costs are still technical challenges that need to be tackled. Fifth, how to achieve mass production of customized flexible ZIBs is still a technical challenge. 3D printing technology has a high degree of freedom and customization range, but the production of customized ZIBs requires long processing (printing, curing) and the cost is prohibitive. Combined with the high safety of ZIBs, 3D printed flexible and customizable ZIBs will generate huge commercial value in smart wearable devices and other fields in the future.

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CONFLICT OF INTEREST STATEMENT

The authors declare no conflicts of interest.

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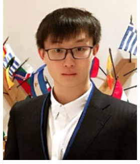

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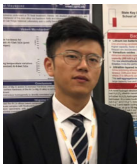

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