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Review

3D Printing of MXenes-Based Electrodes for Energy Storage Applications

Aadharshini Ganesh [1, †], Nisha Gupta [1, 2, †], Prosenjit Saha [2], Pallab Bhattacharya [1, *]

1. Functional Materials Group, Advanced Materials & Processes (AMP) Division, CSIR-National Metallurgical Laboratory (NML), Burmamines, East Singhbhum, Jamshedpur, Jharkhand-831007, India; E-Mails: aadharshini.nitt@gmail.com; guptanisha291995@gmail.com; pallab.b@nmlindia.org
2. Centre for Interdisciplinary Sciences, JIS Institute of Advanced Studies and Research (JISIASR) Kolkata, JIS University, 17th Floor-Arch Water Front Building, Salt Lake City, Kolkata 700091, India; E-Mail: prosenjitpoly@jisiasr.org

† These authors contributed equally to this work.

* **Correspondence:** Pallab Bhattacharya; E-Mail: pallab.b@nmlindia.org

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Abstract

Energy storage devices (ESD) including batteries, and supercapacitors are becoming progressively imperative in the transition to a renewable energy future, as they enable the integration of intermittent renewable sources into the grid and provide backup power during outages. There are already reviews available on various energy storage materials and systems. However, the challenges in the choice of suitable materials and fabrication technology are yet to establish for the commercialization of affordable and efficient ESDs in every aspect of practical needs. Therefore, we realize that the review on the newly developed two-dimensional (2D) MXenes-based energy storage electrodes and devices fabricated through suitably advanced 3D printing technology is the need of the hour, and will be able to attract broad audiences of the related field. MXenes are a class of 2D materials having lamella structures that have shown great promise for energy storage applications due to their versatile redox behavior, high surface area, high electrical conductivity, and ability to accommodate intercalated ions. However, the processing of 2D MXenes suffers from serious agglomeration due to weak Van der Waals attraction and reduces its actual energy storage performances. In a few recent studies, it is observed that advanced 3D printing has enabled the fabrication of MXenes with complex and customized geometries, opening up new possibilities for developing high-performance energy storage devices. Therefore, this review is important for a comprehensive discussion on this topic. So, in this review, we discuss the recent breakthroughs in 3D printed MXene-based batteries and supercapacitors, the advantages of using 3D printing for the fabrication of tailor-designed MXenes-based ESDs, existing challenges, and the opportunities available for further exploration towards the successful commercialization of ESDs. Overall, this review is an insightful articulation for the future seeking to stay at the forefront of this exciting and rapidly-expanding field.

Keywords

MXene; 3D printing; electrodes; charge storage; supercapacitors; batteries

1. Introduction

Rising pollution and the limited stock of fossil fuels are the two major driving forces that create enormous interest in using clean and renewable energies such as solar, wind, tidal etc. However, renewable energy sources such as sun, wind, and tidal wave are non-transportable, so they are not very suitable for on-demand uses. This is where energy storage devices are important and indispensable. They serve to convert non-dispatchable energy technology into dispatchable energy reserves and boost renewable energy integration, grid stability, transportation systems, and industrial applications [1, 2]. However, the existing technologies are not yet fully-proof to use in every required condition. For example, batteries generally suffer from low power density, short cycle life, and poor safety issues. In contrast, supercapacitors have little energy density but are comparatively more safe and long-lasting. Therefore, continued research efforts are seen on the development of energy storage materials and technologies, and thus, timely review of the energy storage systems is also highly essential.

The selection of appropriate materials and fabrication technology are two critical parameters that facilitate the commercialization of pragmatic, economical, and efficient ESDs in all aspects of real-world needs. In the case of materials, there are several attempts made to search the best electrode materials including activated carbon, graphene, carbon nanotube, transition metal chalcogenides, conducting polymers, metal layered double hydroxides, transition metal oxides or hydroxides, composites of hierarchical design and many more [3, 4]. MXenes are also a class of 2D material that has recently shown great promise as the electrode materials for supercapacitors and batteries. Therefore, in this review, we will discuss mostly the MXenes-based energy storage systems. MXene family comprises two-dimensional transition metal nitrides/carbides with arbitrarily dispersed functional groups over the surface, the formula being $M_{n+1}X_nT_x$, which are acquired by selective etching of A elements from ternary MAX phases ($M_{n+1}AX_n$), where M denotes an early transition metal on the left side of the periodic table (Sc, Cr, Ti, Zr, Nb, V, HF, Mo or Ta), A belongs to group IIIA or IVA elements, X is nitrogen or carbon or both, n implies some whole number between 1 and 4, and Tx denotes functional groups that get struck to the surface [5, 6]. Mxenes have unique properties including high electrical conductivity, hydrophilicity, adjustable interlayer spacing, and tuneable functionalities that make them highly attractive for use in ESDs [7]. However, likely to other 2D materials, MXenes are also highly prone to agglomeration which limits the diffusion of electrolytes and surface area; they also get readily oxidized at higher anode potentials, further reducing the cycle efficiency and lifetime. Recently, to address such issues, the rational design and construction of MXene-based electrodes via 3D printing technology (a digital model-based manufacturing technology that controls product designing, reproducibility, and scalability) are deemed an effective approach. [8-11], and thus this review focuses on the critical analysis of various 3D-printed MXenes-based electrodes and ESDs in detail to attract the attention of the broad audience working in the field of energy materials.

2. Principles and Methods of 3D Printing

The general principles of 3D printing can be summarised as follows:

(i) Designing the 3D model: The first step in the 3D printing process is to create or obtain a 3D model of the object you want to print. This can be done using 3D modeling software or by scanning an existing object using a 3D scanner.

(ii) Preparing the model: The 3D model is then prepared for printing by converting it into a format that3D printer can read. This process involves slicing the model into thin layers and generating a set of instructions for the printer to follow.

(iii) Loading the material: The next step is to load the material that will be used to create the object into the 3D printer. The most commonly used materials for 3D printing are plastics, metals, ceramics, and even food can also be used.

(iv) Printing the object: Once the material is loaded, the 3D printer begins printing. The printer heats or melts the material and then deposits it onto the print bed layer by layer, following the instructions generated in the previous step. As each layer is printed, it is cooled and solidified before the next layer is added.

(v) Post-processing: After the object is printed, it may require some post-processing, such as removing any support structures or cleaning up rough edges. Depending on the material used and the desired finish, the object may also require additional finishing steps such as sanding, painting, or polishing.

The basic methods involved in 3D printing are as follows: Direct ink writing, Inkjet printing, Stereolithography, Selective laser sintering, Selective Laser Melting, Fused Deposition Modelling, Electronic Beam Melting, Laminated Object Manufacturing, ultrasonic consolidation, etc., In general, these are classified into three groups including Liquid-based processes (e.g., Stereolithography, Solid ground curing); Solidly based processes (e.g., Laminated object molding and Fused deposition mold); Powder based processes (e.g., Selective laser sintering) [12, 13]. Material choice and properties affect the 3D printing process. Hence, the material chosen must be on par with multiple design specifications, especially for applications in energy storage systems that deploy conductors, semiconductors and insulators. Numerous materials including metals (Li, Na, K, etc.,) and their oxides, polymers, composites, ceramics, graphene, etc., are extensively used in 3D printing for energy storage applications [14, 15]. Apart from these newly emerging materials called smart materials including shape memory materials/alloys, photoactive materials, piezoelectric materials, chromoactive materials, hydrogels, etc., are opening new doors in the context of energy storage devices which is mentioned in the paper [16, 17]. Because of its unusual features such as high negative zeta-potential, strong electrical and thermal conductivity, and mechanical qualities akin to the parent transition metal nitrides/carbides, MXenes have evolved as a wonder material that offers novel possibilities. The properties of MXenes can be applied to a wide range of societal objectives and demands, particularly storage and energy conversion. In addition to improved electric conductivity and enhanced surface area, MXene possesses rich surface chemistry and other outstanding properties due to the terminal surface groups (-OH, -O, -F). As an outcome of these considerations, several more virtues of MXenes have developed, making them appropriate for a wider variety of applications such as ESDs, dual response surfaces, methane storage, transparent conductor, electromagnetic interface shielding, catalyst, filler in polymeric composite, ceramic, hydrogen storage, hydrogen evolution, and nanoscale superconductivity, to name a few. This is extensively mentioned in these research articles [18, 19].

3d printing has transformed material building and has emerged as a promising strategy for intricate electrode assembly in supercapacitors and batteries. They offer distinctive benefits unlike conventional processes as shown in Figure 1 above. First, a single 3D printing machine can rapidly and repeatedly produce electrodes, current collectors, electrolytes, packaging materials, and other items [20, 21]. Second, by altering the printed ink and the pre-programmed printing, it is possible to adjust the shapes and microstructures of electrodes, current collectors, electrolytes, and packaging materials in the 3D printing process, which can supplement the overall electrochemical performance of devices [22]. Thirdly, 3D printing technologies allow for exact manipulation of the primary factors affecting structural electrodes or ESDs, such as the microstructure, energy and power density, and the areal load in the case of active material.

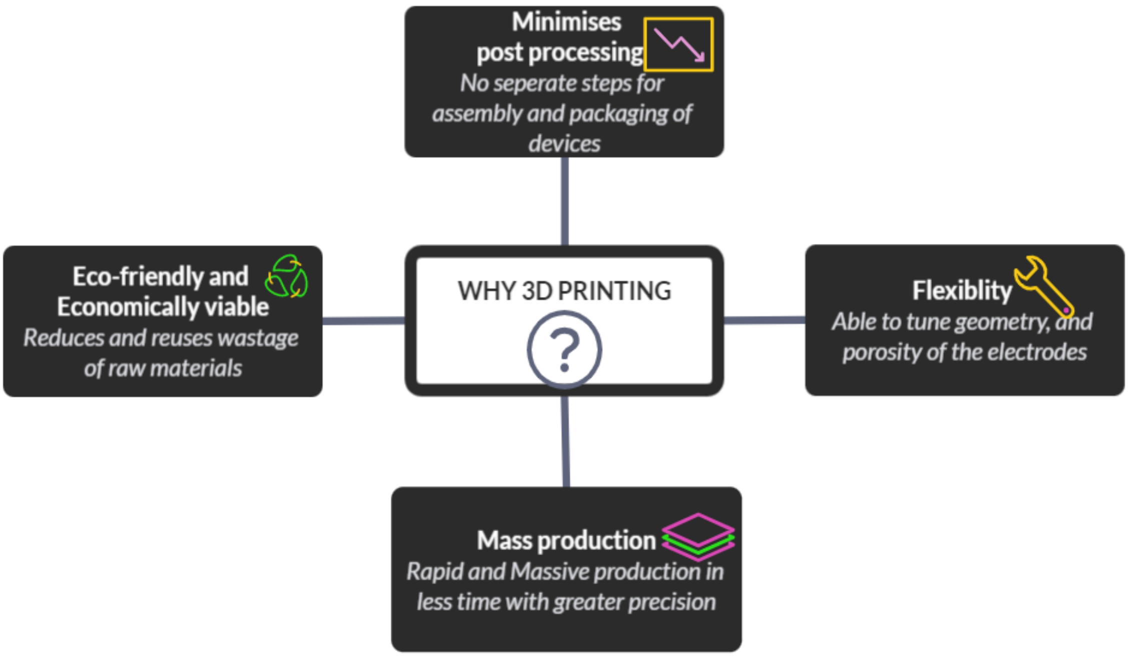

Figure 1 Properties offered by 3D printed ESDs (Energy storage devices).

Direct writing and Inkjet printing are the two most often utilized 3D printing processes for energy storage (ES). Both techniques can easily expand or utilize the conventional ink-like material created by the dispersion of electrode-active chemicals in a solvent. To be appropriately adapted, other 3D printing techniques may require specialized materials engineering and/or process fusion. The following sections give specifics on each process in the ES.

2.1 Inkjet Printing (IJP)

Inkjet printing is a material-saving deposition procedure particularly deployed for liquid-phase materials. These compounds, or inks, comprise a solute dissolved or otherwise distributed in a solvent. The procedure entails ejecting a fixed amount of ink from a nozzle into a chamber by a rapid, quasi-adiabatic reduction in chamber volume via piezoelectric action. In reaction to the imposition of an external voltage, a liquid-filled chamber contracts. This abrupt decrease creates a shockwave in the liquid, causing a liquid drop to discharge from the nozzle [23-28]. The IJP-realized electrodes' thin film properties and higher porosity allow for a higher rate capability [29, 30]. Lithium-sulphur batteries (Li-S) are also recently been created through inkjet printing. The manufactured battery had the largest capacity of any battery manufactured by inkjet printing to date, i.e., 850 $mAhg^{-1}$ sulfur [31].

2.2 Direct Ink Writing (DIW)

It's based on the principle of ink extrusion that behaves as a paste under shear-thinning conditions, i.e., the inks should exhibit ideal rheological behavior and behave as a non-Newtonian fluid. In DIW, the ink or paste is typically composed of a mixture of a material to be printed, a solvent or carrier fluid, and other additives such as fillers, dyes, or crosslinking agents. The ink is extruded from a nozzle under pressure and is typically deposited in a continuous, ribbon-like strand that can be controlled in terms of its width, height, and shape. The ink is then solidified through various means such as solvent evaporation, thermal curing, or UV radiation, depending on the specific ink formulation and application [32, 33].

2.3 Freeze Nano Printing (FNP)

Freeze nano printing is a nanofabrication technique that uses cryogenic temperatures to create complex nanostructures with high precision and accuracy. This technique involves depositing a material onto a substrate in a controlled manner, and then rapidly freezing the material to form a solid, crystalline structure. The frozen structure is then exposed to an ion beam or other etching process, which selectively removes the material to create the desired pattern or structure. Nowadays, complicated structured graphene aerogels (GA) have been defined by combining IJP and DIW techniques with the traditional freeze casting approach, named Freeze Nano Printing (FNP) or ice templating, and is used for synthesizing aerogel [34]. Because of the improvised material loading with a hierarchical porous structure and high surface area, the FNP process offers considerable potential to obtain more specific power and high specific energy [35].

2.4 Stereolithography (SLA)

Stereolithography (SLA) uses a laser to solidify a liquid photopolymer resin layer by layer to create a 3D object. In SLA, a vat of liquid photopolymer resin is selectively exposed to a laser beam, which causes the resin to solidify and bond to the previous layer. The build platform is then lowered slightly and another resin layer is added to the previous layer. This process is repeated until the 3D object is complete [36]. Various printed ceramic/polymer dielectric capacitors are demonstrated by combining a traditional tape-casting ceramic manufacturing method with the stereolithography 3D printing [37, 38].

2.5 Fused Deposition Modeling (FDM)

Fused Deposition Modeling (FDM) involves using a thermoplastic filament, which is heated to its melting point and extruded through a nozzle. The nozzle moves along a predefined path, depositing the molten material to build a 3D object. Here, two extrusion heads normally work in tandem. One is to extrude thermoplastic materials, while the other is to extrude temporary supporting substrates for porous components. The printing materials used in the FDM method must be fluid at high temperatures and capable of efficiently transferring heat. Poly Lactic Acid (PLA), and Acrylonitrile Butadiene Styrene (ABS) filaments are frequently utilized here. Active materials with good conductivity must be added to the PLA or ABS matrix to employ FDM printing for electrodes [39].

2.6 Binder Jetting (3DP)

Binder jetting involves selectively depositing a liquid binder onto a powder bed to create a solid part. The process typically involves spreading a thin layer of powder onto a build platform and then selectively depositing the binder onto the powder using inkjet-like print heads. The binder binds the powder particles together. After printing, the part is typically sintered in a furnace to fuse the powder particles and remove any remaining binder. After completion, it is possible to obtain porous structured plaster-based objects. To create a composite, a second-phase material can be injected into the weak green portion [40, 41].

2.7 Laminated Object Manufacturing (LOM)

In LOM, a sheet of material (usually paper, plastic, or metal) is coated with an adhesive and then cut into the desired shape using a laser, knife, or other cutting tool. A laminating roller is then used in the LOM approach of 3D printing to successfully weld or bond layers of sticky metal, paper, and plastic laminates together. Wire-electrical discharge machining (WEDM) was used to cut Copper foils of 100-m-thickness to fabricate a 3D model cross-section of the electrode. The 3D micro-electrode was then produced by stacking these 2D slices together using vacuum pressure thermal diffusion welding [42].

3. 3D Printed MXenes for Energy Storage

MXene is becoming a promising two-dimensional material (2DM) for energy storage systems. MXene's pseudocapacitive charge storage technology with electric double-layer behavior has enhanced the efficiency of supercapacitors. Moreover, MXene has helped batteries obtain high capacity simultaneously offering rapid charge/discharge due to its apt and unique chemistry and spacing between its interlayer. Such achievements result from MXene's inherent qualities such as excellent electrical conductivity, a specified layered structure, and the capability to withstand modifications, customizing the electrodes to a specific function.

3.1 Historical Progress and Scope of Advancements of MXene-Based Energy Storage Systems

MXene possesses a lesser specific surface area than graphene, which may theoretically increase its energy density and specific capacitance if raised to graphene's level in supercapacitors. To overcome the limitations of the inflexible nanosheets of MXene, this might be accomplished by implementing novel techniques such as intense acid-base treatments to induce porosity within the electrodes. This might also make it possible to host a wider variety of pseudocapacitive nanoparticles and incorporate the appropriate ions into high-voltage electrolytes, enhancing its energy density even more.

MXene is becoming a promising two-dimensional material (2DM) for energy storage systems. MXene has helped batteries obtain high capacity simultaneously offering rapid charge/discharge due to its apt and unique chemistry and spacing between its interlayer. Such achievements result from MXene's inherent qualities such as excellent electrical conductivity, a specified layered structure, and the capability to withstand modifications, customizing the electrodes to a specific function. Thus, MXene-based supercapacitors and batteries manufactured via 3D printing (i.e., the combo of innovative materials and fabrication techniques) have advanced beyond typical button cells to more flexible alternatives [43-47]. To keep up with the recent trend of electronic equipment filling spaces compactly, supercapacitors, miniature devices that store energy are essential. Comparing MXene to its forerunner graphene, quick advancements have already occurred in improving its volumetric capacitance. However, it can be enhanced even further by enhancing both the chemical and the physical elements [44]. Technically, MXene is a promising Lithium-air batteries cathode. However, there haven't yet been any studies that appear to have taken advantage of these conceptual recommendations. The following summarizes a few broad principles for developing desirable MXene-based materials for energy storage applications. However, it needs serious attention though in the dimensions like –

(i) MXenes for flexible electrodes: Near future will be a demand to put pouch-style supercapacitors and batteries together and connect them with wearable or flexible gadgets. Thus to achieve this, we should increase our stockpile of solid-state electrolytes [46].

(ii) Low-Cost and High-Efficiency Preparation: Large-scale MXene preparation should be done under benign reaction conditions, like ambient air, a least corrosive, nil toxic etchant, and room temperature.

(iii) Precise Prognoses: The chemistry and degradation processes in MXene-based energy storage devices are much more complicated for which built-in machine learning models are imperative.

(iv) Reliable chemical stability: Aqueous suspensions of single- or few-layer Ti_3C_2 sheets established by the reconfigured Ti_3AlC_2 exhibit a longer shelf life. These developments offer new approaches to designing highly stable MXene. Due to MXene's ease of oxidation into TiO_2 by oxygen, significant reconstruction and degradation result. Extensive research is still needed to control MXene's chemical stability.

(v) Enhanced surface area: A larger specific surface area could more easily enable adequate electrolyte infiltration and shorten the ion diffusion length. The specific surface area of MXene (~100 m^2g^{-1}) should be enhanced to match the rate capability requirements, opening the door for practical applications.

(vi) Improved Interaction with Interfaces: During cycling, the weakly physically coupled MXene and nanomaterials can produce significant resistance, pulverization, exfoliation, and aggregation which can be minimized by stronger interfacial bonding techniques in the MXene-based nanocomposites.

(vii) Condition for Actual Test: Research on MXene-based complete cells, which calls for substantially higher current densities and greater areal mass loadings, has been comparatively underrepresented up to this point. Thus, full-cell electrochemical data are the need of the hour for the future development of grid energy storage devices.

(viii) Upgraded applications: Other MXene, including those based on vanadium (V) and niobium (Nb), have recently shown special features and uses. The MXene above and MXene-based materials for additional new energy applications, such as aluminum-ion batteries, potassium-oxygen batteries, calcium-ion batteries, lithium-carbon dioxide batteries, magnesium-ion batteries, calcium-oxygen batteries, etc., require to be carried out in order to further address contemporaneous environmental and energy challenges [47].

3.2 3D Printed MXenes Based Supercapacitors and Their Performances

This has covered a broad range, from typical rigid sandwich-type supercapacitors to show great-volumetric capacitance and study mechanisms to super flexible interdigital MSCs to show the potential of MXene in supporting microscopic electronics with varied form factors. Because dissimilar intrinsic qualities might improve a particular characteristic, active supercapacitors materials are chosen depending on their intended purpose. For example, a material will significantly increase the electrical double-layer capacitance if it has a huge active surface area accessible to electrolyte ions; typically, this applies to carbon-based materials such as graphene. This is insufficient to meet the needs of emerging microelectronics, which need more energy generation from a smaller footprint. In order to maximize the energy and power density of supercapacitors, transition metal oxides and transition metal dichalcogenides are added [48-52]. However, to boost their performance rate, they require the usage of binders derived from carbon or current collectors due to the low electrical conductance of carbon. In this sense, MXene has become a fantastic choice to address this quandary.

An MXene-reduced graphene oxide composite sans additives that exhibit excellently resists deterioration under more than 250 percent stresses in uni-axial or bi-axial directions was created. Yet after 1000 cycles, a reasonable capacitance of 19 $mFcm^{-2}$ may be well maintained at almost 100 percent. This stretchability is an essential component of wearable MSCs since it guarantees resistance to damage. However, self-healing, which elevates damage recuperation, is the final test for all. Though wearable systems are still far from being able to use self-healing supercapacitors that are now available, the display of functional prototypes is stunning [52]. The research article [48], demonstrates 3D printed MXene ($Ti_3C_2T_x$) manufactured by continuous extrusion-based technique, i.e., direct ink writing helps produce current-collector free supercapacitor. Such a 3D-printed device has a remarkable areal capacitance of 2.1 Fcm^{-2} at 2 $mAcm^{-2}$ and a gravimetric capacitance of 242

Fg^{-1} at 0.2 Ag^{-1} with a capacitance's retention of more than 90% after 10,000 cycles. It also has a higher energy density of 0.025 $mWhcm^{-2}$ and a power density of 0.6 $mWcm^{-2}$ at 4.3 $mAcm^{-2}$. The self-sustaining printing and designing technique established in this work is expected to apply to the fabrication of improved performance customized multidimensional and multiscale architectures of structural and functional materials for integrated devices in a broad range of purposes and uses. The customizable direct ink writing technology demonstrates the prospect of additive-free MXene inks for expandable manufacturing of incredibly simple printable electronics components including conductive tracks, micro-supercapacitors, and ohmic resistors. This developed 3D printing method is suitable for producing versatile MSCs on paper and polymer substrates. Those printed solid-state systems exhibit remarkable electrochemical performance, with areal capacitances of up to 1035 $mFcm^{-2}$ [49]. Another study [50] identified that Nitrogen doping improves MXene's electrochemical performance by increasing redox activity and conductivity. As a result, MXene-N inks are made by optimizing ink viscosity to suit 3D extrusion printing. Interestingly, the 3D-printed MXene-N-based supercapacitor possesses an areal capacitance of 8 Fcm^{-2} for electrodes with three layers and a substantial areal energy density of 0.4 $mWhcm^{-2}$. Another paper [51] demonstrates that asymmetric supercapacitors (ASCs) are constructed by using exfoliated $Ti_3C_2T_x$ (Ex-$Ti_3C_2T_x$) negative electrode and transition metal chalcogenide (MoS_{3-x}) coated 3D-printed nanocarbon template (MoS_{3-x}@3DnCF) as the positive electrode in H_2SO_4 or Polyvinyl Alcohol gel (PVA) electrolyte with a broad range ΔV of 1.6 V. In Figure 2a the schematic diagram of 3D printed micro-supercapacitors (MSCs) with interdigital architecture was manufactured using the direct Ink writing technique. Figure 2b gives an idea about the layered structure of the printed MXene flakes, which are horizontally aligned. Figure 2c represents the flexibility of the device by changing bending angles. There is little significant change in the rate capability, conductivity, adhesion, and specific capacitance [49]. In Figure 2d the schematic representation of 3D printed MXene for MSCs devices was designed using an extrusion-based direct ink writing technique. Figure 2e shows that along the temperature gradient ice crystal grow unidirectional where the MXene flakes are well aligned vertically to form the ordered channels. The cycling performance of the developed 3D printed MXene hydrogel (Figure 2f) at 100 mVs^{-1} shows a capacitance retention of 95.5% after 10000 cycles and in the inset image the CV curve of MXene hydrogel initial 1^{st} cycle and after 10000 cycles, indicates the good cycling stability of as-prepared MXene hydrogel [52].

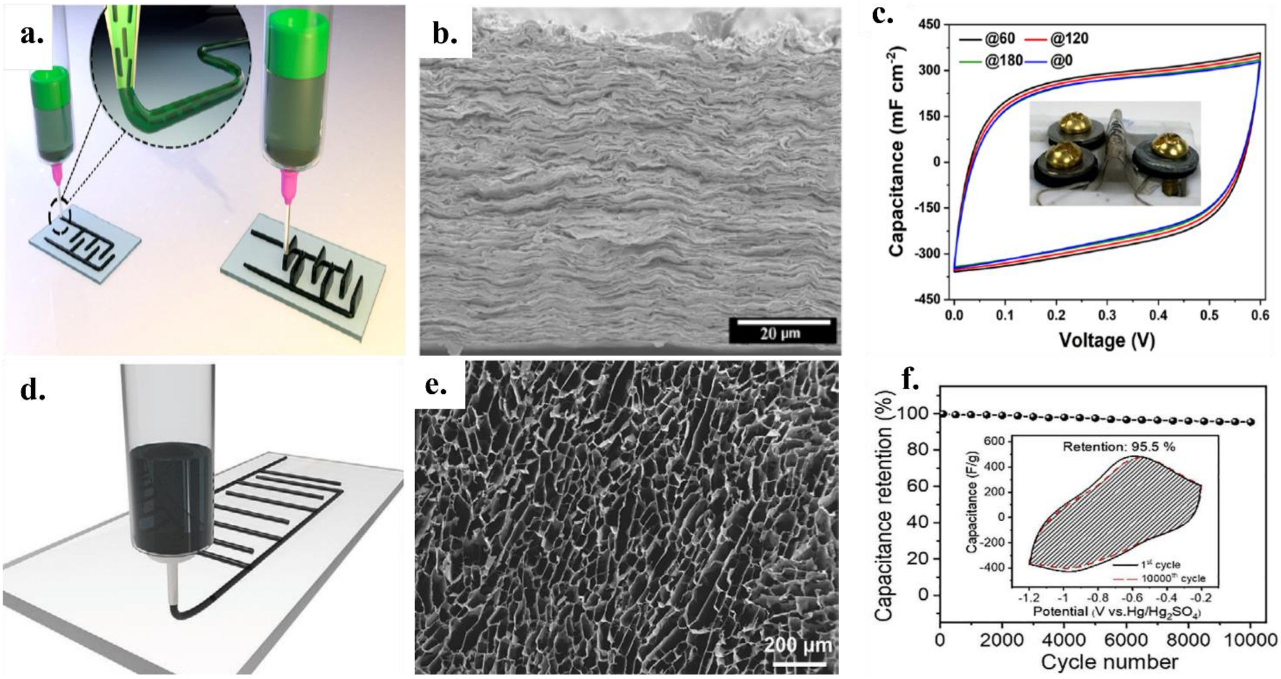

Figure 2 a) Diagram exhibiting 3D printing of MSCs with interdigitated topologies. The flakes are aligned horizontally in the direction of nozzle movement due to the shear tension created in the nozzle. b) Cross-sectional SEM images of the electrodes in the MSC-10 device at various magnifications demonstrating that $T_{i3}C_2T_x$ flakes are densely piled up and laterally positioned; c) Electrochemical performance of MSCF-1 at a 10 m V s^{-1} scan rate under various bending angles (starting with 60°, succeeded by 120°, 180°, and 0°). (Copyright with permission of ACS nano 2020) [49]; d) 3D printed MXene flakes; e) SEM images of top-view ordered MXene hydrogel; f) Cycling performance of ordered MXene hydrogel. Inset: comparison of the CV curves before and after 10000 cycles (100 mVs^{-1}) (Copyright with permission of Wiley 2021) [52].

Additionally, MXene has hit the market as a competing active substance that works with various self-healing strategies. Encasing a quasi-solid state supercapacitor possessing self-healing polymeric materials, such as carboxylated polyurethane, is a typical self-healing technique. Although the MXene-based composite in question cannot repair on its own after breaking, appropriate alignment may be capable of re-establishing electrical contact (with negligible conductivity loss). The withholding of 81% capacitance shows these approaches have reasonable recovering efficiency. However, the internal resistance's value rises by over 50 ohms throughout five recovery cycles. Therefore, it is much more advantageous to develop inherently self-healing MXene-based electrodes. But to date, no such processes have been identified. However, the development of completely healing supercapacitors made from MXenes would be made possible by recent research on incorporating MXene into self-healing viscoelastic materials and demonstrating their capacitive performance based on dielectric [53].

3.3 3D Printed MXenes Based Lithium Based Batteries and Their Performances MXene

Rechargeable lithium-ion batteries (LIBs) contributed to the necessary but unanticipated growth of electric and hybrid vehicles and mobile devices by minimizing reliance on a permanent power supply. The research on LIBs is primarily concerned with keeping up with the shrinking of electronics by minimizing footprint and improving energy density while charging more promptly [54]. MXene, which promises to expand into systems other than Li-ion and upgrade LIBs, has quickly entered the race as a powerful contender [55, 56]. Due to its huge weak interfacial forces, specific surface area, open structure, and surface functional groups, MXene was examined as a LIB anode material and was discovered to produce promising outcomes. Due to the huge number of combinations and configurations that may be made with MXene, which comprises of M early transition metals, X (C or N or both), and T (functional groups – O, OH, F), the functionality can be adjusted. In particular, Ti_2C, Nb_2C, and V_2C are the chronology of the MXenes' particular capacities, but Nb_2C and V_2C exhibit excellent rate capabilities. Additionally, the voltage waveforms of various MXenes differ greatly, indicating the applicability of various MXenes in anodes or cathodes. Nevertheless, the band structures of the functional groups frequently differ substantially. For instance, although functionalized MXene is a semiconductor, bare MXene is a magnetic metal. They also have an impact on Li absorption and transportation. By widening the gap between layers, non-native functional groups including chlorides would benefit MXenes sterically and counteract the negative impacts due to the ingrained OH, O, and F functional groups. In fact, because of the changing degree of surface characteristics, perhaps the stoichiometric ratio of 'M' derived from the same species synthesized using different processing technologies might potentially influence the performance of LIBs. The incorporation of Li ions is typically hampered by Ti_2CT_x because it generally has a lower c-lattice value than $Ti_3C_2T_x$ [57].

The possibilities for rendering metal-based Lithium batteries safe and secure through mechanism analysis, enhancers, and host substrates in light of the enormous potential available is of primary concern. Interestingly, MXene has also contributed to this field by serving as a powerful 3D lipophilic host [58-63]. However, at high Lithium loading of 92%, the MXene-based 3D model enables uniform lithium diffusion that impressively inhibits dendrite development while maintaining a high current density. Consequently, 112 days of operation at 0.5 $mAhcm^2$ were completed with 99% coulombic efficacy. The secret is to use a 3D structure to homogenize dendrite proliferation to establish a stable solid-electrolyte interphase or to uniformly nucleate Li-dendrites over the host utilizing methods such as nanoparticle doping. MXene, which has high conductivity, wide surface area, and lipophilic characteristics, is particularly advantageous as a platform for such a nucleation method. This ensures that lithium dendrites will develop in a defined way, forming bowl-shaped structures, and preventing unlimited expansion of volume while maintaining a larger volumetric capacity of 40 mAh [64]. The LiS battery has emerged as an appealing upcoming approach for sophisticated energy storage. The actual deployment of Li-S batteries has also been impeded by unmanageable dendritic growth at the anode and poor cathode loading performance. To overcome such challenges, the following articles propose 3D printed Li-S fuel cells (3DP N-p$Ti_3C_2T_x$/S||3DP N-p$Ti_3C_2T_x$@Li) that can keep functioning for 250 cycles with a sulfur loading of 7.6 $mgcm^{-2}$, with a capacity decay of 0.06% each cycle. Astonishingly, after 60 cycles at 12 $mgcm^{-2}$, an ultimate capacity of 8.5 $mAhcm^{-2}$ is obtained [53]. In situ fabrication of MO_x-MXene (M: Ti/V/Nb) heterostructures as bulky and versatile hosts to obtain excellent battery performance with synchronized polysulfide immobilizations and conversions. Mathematical calculation shows that the real implantation of oxides improves the kinetics of polysulphide transformations without compromising MXene's inherent conductivity. Thus, the typical VO_x-V_2C/S electrode has an improved volumetric capacity (1646 $mAhcm^{-3}$ at 0.2 C) and cycling stability (631.2 $mAhcm^{-3}$ following 1500 cycles at 2.0 C with a potential decline of 0.03%

each cycle). More appreciatively, 3Dprinted sulfur electrodes based on VO_x-V_2C host easily capture an areal capacity of 9.74 mAhcm^{-2} at 0.05 C with an enhanced sulfur loading of 10.8 mg cm^2, offering prospects for the creation of sensible Lithium Sulphur battery [54].

3.4 3D Printed MXenes Based Batteries Other Than Lithium-Based Batteries

Despite having larger ionic radii than Lithium, these ions nonetheless show superior rate capability, probably caused by the MXene's increased interlayer spacing and excellent ionic and electronic conductivity. Mechanically, MXene can withstand the strain caused by the additional nanoparticles' volume expansion and precludes their pulverization following de-sedation. In contrast, the nanoparticles lower the surface energy and inhibit MXene from stacking again. Digitally, the nanoparticles might help to improve the valence band and conduction band overlapping, facilitating the donation of electrons by MXene [65]. MXene usually conducts electricity, but its ionic equivalent is more fascinating since it shows hopping ion(s) of Sodium, Potassium at the top Titanium and Carbon sites. The monolayer containing MXene slides horizontally due to the process that permits ions with multivalency like Al^{3+} to have lower diffusion barriers. Hybridization of MXene with other nanomaterials, such as transition metal oxides or black phosphorus, could further lower the diffusion barrier, at least for the Na^+ ion, improving stability and specific capacity [66].

Concerning the context of Figure 3, to capture higher energy and power density, Sodium-Ion Hybrid Capacitors (SICs) devices based on 3D printing of current-collector-free N-$Ti_3C_2T_x$ anode along with AC cathode can be used and whose ink manufactured as in Figure 3a. The porous N-$Ti_3C_2T_x$ material, with its porous opening structure and homogenous nitrogen doping, facilitates rapid electron/ion routes and electrochemical reaction kinetics, leading to improved sodium-ion retention behaviour. As a result, the thus obtained SIC full-cell has a superior gravimetric energy/power density of ~101 Whkg^{-1}/3269 Wkg^{-1} and a favorable areal energy/power density of ~1.2 mWhcm^{-2}/40.2 mWcm^{-2}. This study presents a viable method for fabricating and developing energy storage devices with high power and energy density [54]. A 3D-printed sodium-ion hybrid capacitor (SIC) based on an activated carbon cathode and nitrogen-doped MXene (N-$Ti_3C_2T_x$) anode is illustrated in Figure 3b. It also shows that SIC-4 exhibits good cyclic stability with a capacity retention of 75% over 3500 cycles at 2 Ag^{-1}. A sacrificial template approach can produce N-$Ti_3C_2T_x$ with a well-defined porosity structure and consistent nitrogen doping [67].

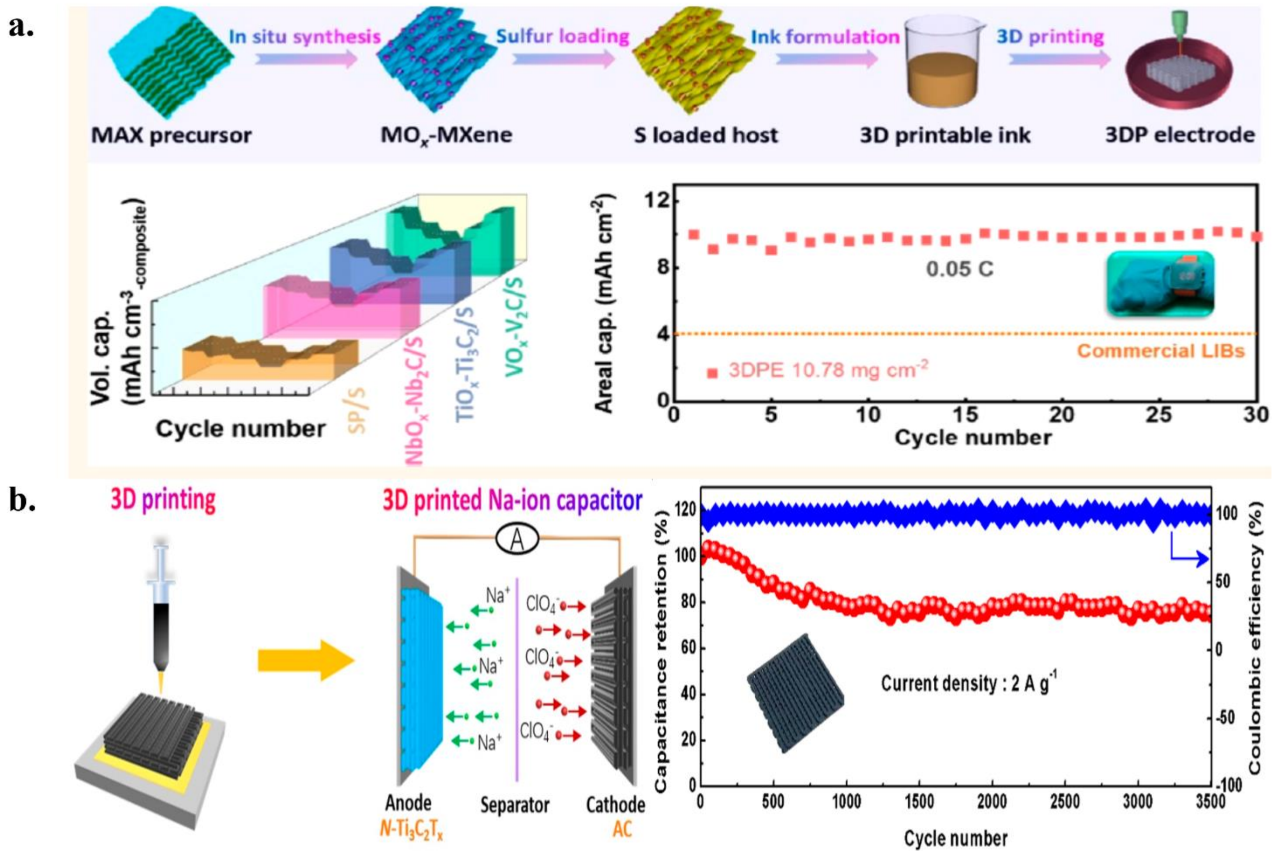

Figure 3 a) MOx-MXene (M: Ti/V/Nb) heterostructure formulation for 3D printing of sulphur electrodes (Pictorial depiction of the 3DP MO_x-MXene/S electrode fabrication techniques.) (Copyright with permission of ACS nano, 2020) [54]; b) 3D porous N doped $Ti_3C_2T_x$ MXene and 3D-printed SIC (SIC-4) cycling performance at 2 Ag^{-1} with inset showing a 3D-printed woodpile N-$Ti_3C_2T_x$ electrode in actual situations. (copyright with permission of ACS, 2020) [67].

Batteries made of multivalent ion(s), like Mg^{2+}, Ca^{2+}, Al^{3+}, and Zn^{2+}, generally have remained slow to catch on than K^+ and Na^+ ion batteries. Modern V_2CT_x MXene-based rechargeable Aluminium ion batteries have shown greater specific capacities over 300 $mAhg^{-1}$ at higher current densities of 100 mAg^{-1}, however a reason for the potential loss that occurs continuously when cycling has not yet been discovered. Considering capacity retention rising from 26.5 percent to 72.3 percent after 120 cycles, the surface morphology of $Ti_3C_2T_x$ MXene with certain other nanoparticles like TiO_2 and sulfur has demonstrated to rise its stability. After 120 cycles, the discharge capacity too increased [68]. Earlier research on multivalent ion batteries based on MXene revealed a high dependence on ion intercalation as the principal energy storage method. They all suffered from dramatic capacity loss after just a few thousand cycles. However, the latest review on zinc ion batteries developed on V_2CT_x showed a startling increase in capacity reaching 508 $mAhg^{-1}$ across 18 thousand cycles [69].

4. Challenges in Manufacturing Via 3D Printing

4.1 Optimisation of Shape

Design space optimization might significantly affect component fabrication, resulting in cheaper production costs by reducing the need for raw materials, manufacturing time, electricity requirements, and atmospheric impacts [70]. It is not an easy challenge to significantly improve the design space. The goal is to determine the most appropriate method for filling the design space with substances that maximizes the number of design factors, including weight, strength, and area. The size and degree to which the search space in the design space for maximum dispersion depends on the component and their attributes under consideration. Even if a suitable arrangement is discovered, it might not work among all 3D printing techniques. Problems may result from materials, walls that have not been constructed well, or inadequate support [71].

4.2 Pattern Making for AM

By involving 3D printing, complicated configurations that cannot be produced by other production processes like machining, milling or molds can be created. The capacity to incorporate hierarchical complexity within components, the addition of numerous materials to a single component, and completely functional integrated mechanisms seem to be advantages of employing 3D printers. However, to take perks of the special features of additive manufacturing, the schematic design should be reconsidered in light of conventional methods, and newer tools must be developed to support such a design type [72].

4.3 Pre- and Post-Processing

The ideal printed component doesn't come from a 3D model right away. Before sending the model to the printers as a set of guidelines for building the component, it must be pre-processed. Depending on the manufacturing method, greater care might be necessary to remove the supporters, enhance the surface finish, or determine specific features or faults after manufacturing the object. Pre- and post-treatment each provide unique challenges that influence how we manage the printing process as a whole [73].

4.4 Methods Used for Printing

There are numerous methods by which it is possible to produce objects when using additive manufacturing. Although layered fabrication continues as the most well-liked and thoroughly studied approach, alternative approaches exist. Every construction approach has benefits and drawbacks, similar to all technical disciplines. The physical or mechanical qualities of the item may be significantly affected by which is adopted for processing. The concept of direct synthesis serves as the foundation for each of the methods mentioned [74].

4.5 Controlling Errors

Even 3D printing possesses its set of flaws in the production chain. Because of a lack of quality assurance and control standards, the existing machinery available on the market might not be the most dependable. Three types of faults can be found in 3D printing: errors during data preparation, processing errors, and material problems. Although mistake elimination is possible at the data preparation stage, it could not be possible during the process material error step due to the faults' nature. For the other two groups, error correction is occasionally a more advantageous choice, though a much more challenging one [75].

4.6 Printing of Multi-Material

For homogeneous components, 3D printers would fabricate them from a diverse range of materials. To avail advantage of different qualities; some situations, however, call for using various materials. We commonly refer to a printed product made of multiple materials as a heterogeneous entity. But it's not as easy as simply dumping in more materials. A multi-material object could be divided into two categories in general. Multiple materials may be present in a heterogeneous solid model of the component, but the various materials' segments can be considered discrete regions with rough edges. A gradient among materials with more complicated boundary conditions can be found in functionally graded models [76].

Modeling and production are the main obstacles in printing printing with various materials. Few CAX systems are available right now that can properly handle various materials. There are various suggested approaches for modeling heterogeneous elements, but each approach has advantages and disadvantages of its own. Once the modelling is complete, the printer being used must have appropriate mechanisms for printing with multiple materials. Whether the printer can fabricate such a design, caution must be exercised to ensure that all ingredients work together properly. Multi-material merging or fusing is highly reliant on material science and therefore can present multiple difficulties that must be considered in the production and modeling [77].

4.7 Issues with Maintenance and Hardware

The machines for additive manufacturing that are now available on the market have advanced significantly since their beginnings a few decades back. Despite this, they continue to experience performance and maintenance woes. Each machine must be properly configured with the right specifications for a successful fabrication. Material limitations, energy restrictions, and numerous process-specific regulations are some of the criteria. Even though a setup succeeds with one component, it does not imply that it will be adequate to construct any given random structure. An incorrect setup does not imply that a component will not be printed, whereas it could result in poor dimensions, geometry, or aesthetics. Although machines function autonomously, it is vital to routinely monitor them to ensure that processes maintain the same level of accuracy. In order to maintain the machines' functionality beyond age, clean-up processes are frequently needed once a component has been done. Each of these factors would add much time to the production cycle [78].

4.8 Alignment

Modifying the alignment of the component to increase or decrease one or more manufacturing criteria is the definition of the alignment problem. The STL file or the actual CAD model can be used. Certain attributes may be more crucial than others based on the component's use or intended function. Anisotropic pieces' orientation can impact their mechanical behavior, build time, and quality. Based on the procedure, manufacturing limitations such as supporting or depositing parameters may also have to be considered [79].

4.9 Slicing Problems

A CAD or STL model must be sliced into segments to fabricate the product in all layered manufacturing techniques. To determine the geometry of the slices, the model is dissected using horizontal planes. The thickness of the layer determines the height of the slicing. This slicing procedure prepares the model for designing the deposition path [80]. Nowadays machines most frequently slice using a technique called homogeneous slicing, which ensures that each slice, irrespective of configuration, has the exact layer thickness. These segments are usually called 2.5D contours since they often lack the model's indigenous vertical geometry. For this rationale, there are two fundamental difficulties with the slicing problem. Firstly, the stepped corners are produced by the 2.5D contours, sometimes known as the staircase effect. The latter is the containment problem, which occurs when the slice doesn't exactly match the layout but instead lies outside or within the original model. Both of the above factors contribute to the precision and surface quality issues. The slicing procedure directly impacts the part's building time, accuracy, and smoothness [81].

4.10 Speed

Any process may be constrained by how long it takes to fabricate a component. Comparing 3D printing to conventional production methods is complex. 3D printing is far less difficult to establish than milling and can create more intricate objects with a single print. It is possible to mistakenly think of speed as the height of the component that must be manufactured. Meanwhile, focusing on the build height and throughput could provide a more appropriate statistic because 3D printing methods can potential use the build area for numerous pieces. Although the printing process' main constraint is hardware, pre-processing speed still takes time and is almost entirely determined by software [82].

4.11 Health and Safety Aspects

Safety and health considerations are among the most pressing issues confronting 3D printing. 3D printers emit various harmful super fine particulates and volatile organic compounds (VOCs), which can make users sick with respiratory disorders. The likelihood of 3D printing causing pollution rises as its usage expands. As a result, pollution brought on by 3D printing must be strictly controlled; otherwise, it could spark yet another global problem that will be complex to tackle [83].

4.12 Cybercrime and Copyrights Violations

Prototypes made with 3D printing are essentially digital data, making them susceptible to theft or copying. There haven't been many reports of these copyright infractions, but as technologies evolve more ubiquity, more cases will emerge [84].

4.13 Low Productivity

Another obstacle to the widespread commercialization of 3D printing is that the productivity is still too low to meet the demands of developing technical standards. An additional issue that requires consideration is the fact that AM is only recommended for production on a small scale and has significant production constraints while producing massive quantities of merchandise [85].

5. Summary and Future Perspectives

It is thought that 3D printing technology, which combines advanced manufacturing techniques and computer-aided design to create functioning structures, is a breakthrough and highly alluring method for creating efficient electrochemical energy storage systems. Rapid prototyping and geometric shape design are two areas where 3D printing stands out from traditional production techniques, especially regarding the large surface area and complicated 3-dimensional structure designs. A handful of 3D-printed devices that store energy have recently been disclosed, demonstrating the professional community's significant attention to the technology. Understanding the advantages, and disadvantages and being enriched with the latest developments of 3D-printed energy storage systems are essential for furthering the material design and experiencing technological advancements. So, an overview of the research on current revelations in 3D printing technology for energy storage devices has been discussed above thoroughly which, we believe, will attract a broad audience working in 3D printed energy materials. The architecture of printing media, the procedure for printing, the usage of printed devices, and the difficulties encountered by current printing processes are also discussed, enlightening many new scopes and areas for further developments in the field.

3D-printed MXenes for energy storage applications are also accentuated. There are still many unknowns with MXenes. This includes the magnitude of the family, the maximum theoretical storage capacity for lithium, and other energy-related uses. The MXene family will continue to grow in the future through various elemental combinations, according to theoretical predictions. MXenes' capabilities as rechargeable Li-ion anodes will be boosted by using composites with additional elements such as Si, Ge, Sn, and other transition metal oxides. Additionally, while synthesizing MXene composites with broader band gaps semiconductors like TiO_2 and tunable band gap semiconductors like phosphorene, photocatalytic processes can be tuned. SSo, problems in different forms exist on various MXene-based energy materials which might be solved by using, controlling and optimizing the 3D printing technologies. Upcoming energy storage technology has the potential to be manufactured using one of the most anticipated, promising and potential manufacturing processes, 3D printing, which has the advantages of being cheap, having a rapid construction rate, exactness in positional accuracy, and broader material options.

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Competing Interests

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