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PROGRESS REVIEW

Advancing flexible electronics and additive manufacturing

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There is high interest in the synergism of thin-film and flexible electronics with additive manufacturing. This review primarily focuses on the prospective developments in convergence with flexible electronics manufacturing technologies. Specifically, this paper covers the latest 3D printing and hybrid manufacturing technologies, the utility of specific types of materials, their functionalization and characterization, post-processing and testing strategies toward fabricating robust and application-specific flexible electronics. Besides exploring the advances in this area of research—it also highlights the limitations and gaps that have been observed in the previous years that will challenge and offer opportunities for advancing research and development. Lastly, the future of 3D-printed flexible electronics is discussed in the aspects of customizability, scalability, and its game-changing and state-of-the-art potential for intelligent sensing, instrumentation, and wearables for various medical, engineering, and industrial applications.

1. Introduction

Flexible electronics are a class of electronic devices that commonly have substrates characterized and made of thin or long stretchable materials.[1)] At a certain degree, it can be folded, stretched, elongated, rolled, or bent to either adapt to specific rugged applications or to trigger changes in electrical signals from changes in stimulus like how a sensor works.[2)] The concept of such a class of conformable electronics has existed ever since the first electronic devices were invented. What sets it apart from any other electronics class is its flexibility (more mechanical) when the application calls for it. From when flexible solar cells and thin-film transistors[3)] started gaining ground around the late 1970s to their significant contributions to biosensors in the last decade, the way these are fabricated had to revolutionize the traditional manufacturing processes. Although the actual electronic part itself may still conform to conventional electronic device fabrication methods, the challenge has always been its integration into its flexible substrate.[1)]

Flexible electronics, as it becomes a popular class of the electronics industry, likewise demands "flexible" ways of fabricating them that address the integrative approach to customizability and application-specific needs.[4)] One innovative way to push the limits in manufacturing of flexible electronics has been the use of advanced methods of manufacturing such as 3D printing, wherein it opens a whole new manufacturing process and breed of products, such as wearables,[5)] biosensors,[6–8)] conformal antennas,[9)] and flexible energy harvesters and storage,[10)] that somehow heed the demand for custom-built application-specific devices from the fields of medicine, renewable energy, and engineering. 3D Printing is often synonymous with the term additive manufacturing (AM). In several review papers, we recently reviewed advances in 3D printing methods, materials, including 3D printing for electronics,[10)] mechanical testing,[11)] high-performance polymer composites,[12)] graphene and polyurethane,[13)] stereolithographic apparatus (SLA) 3D printing,[14)] thermoplastic composites,[15)] graphene and SLA,[16)] carbon black and selective laser sintering (SLS),[17)] silicone elastomers,[18)] hydrogels,[19)] and viscoelastic materials.[20)] In this case, AM could refer to a build-up of layered or multi-layered structures that build on multi-materials or a computer-aided design (CAD) framework design origin. 3D printing in flexible electronics could be a combination of CAD-based layer addition and a more serial plug-and-place or pick-and-place design to construct a device.

The purpose of this review is to provide a fresh and concise overview of the recent advances, grand challenges, and prospects in the advanced manufacturing of flexible electronics. This paper mainly covers the processing and functionalization of materials, post-processing and testing, 3D printing technologies and hybrid approaches, the newest family of flexible devices, and sustainability aspects of flexible electronics.

2. Advances in manufacturing flexible electronics

2.1. Processing and functionalization of materials

Recent advances in flexible electronics enabled researchers and electronics industry professionals to develop different processing techniques and functionalization of its materials.

Among the popular processing techniques to fabricate flexible electronics are transfer printing technology (TPT), two-dimensional printing (2D printing), and three-dimensional printing (3D printing). Figure 1. illustrates the

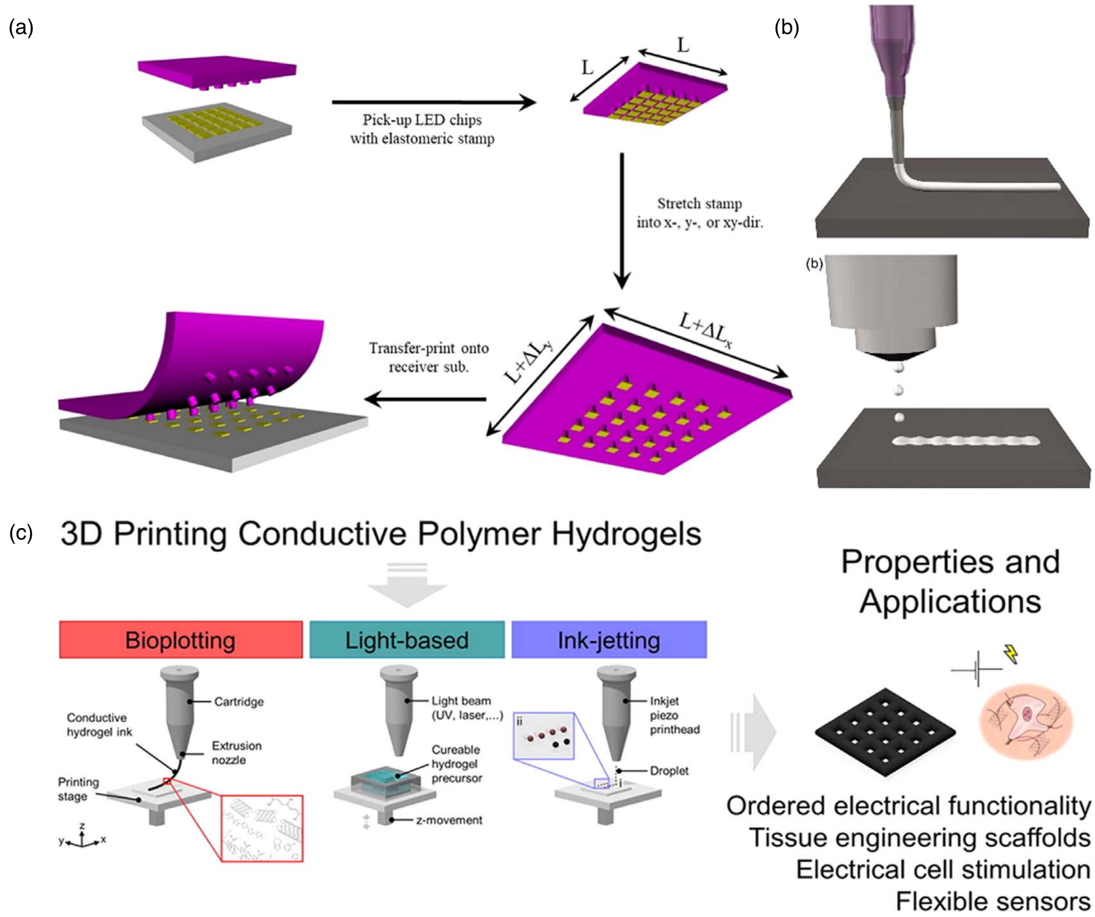

Fig. 1. (Color online) Figure 2.1(a) Schematic illustration of non-deterministic transfer printing of LED chips with controllable pitch using a pillar-patterned, stretchable elastomeric stamp,[24] (b) Schematic view of ink-based deposition schemes: droplet jetting and continuous filament writing,[25] and (c) 3D Printing Conductive Polymer Hydrogels.[26]

difference in the setup for TPT, 2D printing, and 3D printing. TPT is a widely used technique to manufacture flexible electronics through reversible interfacial adhesion of rigid functional devices with elastomeric substrates.[21,22] The 2D printing utilized 2D functional ink or liquid-based/aqueous electrolyte integrated into the system as sensors, biosensors, wearable displays, and other smart applications.[23] For 3D printing flexible electronics, the process involves 3D molded interconnected devices (3D-MID) that require printing of complex 3D structures and substrate geometries.[10]

The process of using 3D printing in flexible electronics is dependent on the complexity of its design and the 3D printing technology that will be utilized. A preset pattern that directly targets the substrate during printing is a complex process involving smart interactive operations between operators and 3D printers.[27] Hoerber et al. demonstrated in their research the enhanced process for building 3D devices by aerosol jet printing and powder bed-based printer. Both methods involve the integration of surface mount technology (SMT) components into the structure. In aerosol jet printing, the SMT components were integrated into the structure via assembly and interconnection. While in a powder bed-based printer, the SMT components were buried into the powder bed with the cavities created during the process.[28]

Surface functionalization of flexible electronics is a method to introduce electrical conductivity and optical properties to attain its intended electronic application.[29] The functionalization of flexible electronics can be done by in situ polymerization,[30] post-polymerization/coating,[31] addition of conductive particles,[32] and ionic conductivity.[33]

2.2. 3D Printing

Numerous studies and review articles demonstrate the applications of 3D-printed flexible electronics. These applications include robots, pumps, wearables, energy absorption, biocompatible materials, and different types of sensors (piezoelectric, piezoresistive, bending sensors, electronic, strain sensors, capacitive force sensors, etc.), and many others.[34] Several examples of these applications that are manufactured using only a single 3D printing process are briefly discussed in this section,[13,35–40] while those with two or more processes (i.e. hybrid)[41–44] are discussed in the next section. Table I summarizes the materials and technologies used in the 3D printing of flexible electronics. It should be mentioned that a key ingredient in flexible electronics is the flexibility of the substrate and therefore the use of materials such as polyurethane (PU), silicone, or polydimethylsiloxane (PDMS), natural rubber, and other thermoset and thermoplastic elastomer compositions.

Christ et al.[35] reported successfully having 3D-printed a flexible and conductive thermoplastic-based strain sensor. They compounded thermoplastic polyurethane with multiwalled carbon nanotube (TPU/MWCNT); filaments were extruded as fused deposition modeling (FDM) was used for the 3D printing of the sensor. The electrical, mechanical, and

Table I. Flexible electronic devices manufactured using different 3D printing technologies and materials.

Material	3D printing technology	Application
Thermoplastic polyurethane/multiwalled carbon nanotube (TPU/MWCNT)[35]	FDM	Sensors
PDMS, TPU, silver micro flakes, NaCl crystals[36]	Extrusion	Stretchable piezoresistive sensor
Rigid polyurethane and flexible Polyurethane[37]	Direct laser synthesis	Bladder pumps, shape-to-fit flexible wearables
polyurethane and a conductive filament[38]	FDM	Multifunctional flexible bending sensors
Conductive filament (Polycaprolactone with carbon black)[39]	FDM	Electronic sensor
Thermoplastic polyurethane (TPU)[40]	FDM	Energy absorption
Thermoplastic polyurethane (TPU)[13]	FDM	Biocompatible materials/biomaterials scaffolds for tissue engineering
Conductive polydimethylsiloxane (PDMS)[41]	Embedded 3D printing (e-3DP)	Strain sensors
Photoresist and oil, with chromium and copper metallic layers[42]	3D Microprinting/direct ink writing (DIW)	3D conductive serpentine microstructures/networks
Acrylonitrile butadiene styrene, conductive electrodes, and dielectric layer[43]	FDM, fiber encapsulation additive manufacturing (FEAM), and thermoplastic elastomer additive manufacturing	Acrylonitrile butadiene styrene, conductive electrodes, and dielectric layer
Tellurium and silver[5]	Aerosol jet printing and extrusion printing	Piezoelectric sensor
Soft materials (multiple materials); PDMS [44]	EMB3D and molding	Soft robot

piezoresistive properties were investigated under monotonous and cyclic loading conditions. MWCNTs increased the stiffness of the TPU for better printability. The elastic modulus slightly decreased, and there was no significant change in the material's conductivity compared with the bulk materials. A high piezoresistivity gauge factor has been measured even under 100% applied strains. Under cyclic loading, a highly repeatable resistance strain response was observed. These characteristics demonstrate the potential applications of such composite material in wearables, soft robotics, and the like. Wei et al. similarly described the preparation of the reprocessable 3D-printed conductive composite foam of polyurethane (carbon black composite) for strain and gas sensing.[45]

Bates et al. successfully 3D-printed hyperelastic cellular arrays, or what they called honeycombs, with graded densities for energy absorption.[40] TPU was used to manufacture hexagonal arrays via FDM. Density grading was done by varying cell wall thickness through the specimen thickness. Samples were subjected to static compression tests where the stress-strain behavior, efficiency-stress behavior, and energy absorption diagrams were obtained. The researchers observed that by grading the density through the specimen thickness, the structures were able to absorb higher total energy under compression loading than the equivalent uniform array. Further, they observed that the graded structures demonstrated higher efficiency at absorbing low energy compression loads than the equivalent uniform array. Lastly, they reported that the continuously graded array had the highest efficiency over a wide range of compression energies. In contrast, the highest peak efficiency was observed in the equivalent uniform density array.

2.3. Post-processing and testing

There are several techniques in post-processing 3D-printed polymers[46] but the most common method for complex 3D-printed structures is the removal of support structures.[47] The support structures are usually made from different materials and can be dissolved from the completed object post-printing.[26] The use of high-resolution printing and lattice structures can minimize the structural support of flexible electronics.[48]

Thermal post-processing is needed to evaporate the solvent in ink. Usually, the process of annealing, laser curing, plasma treatment, and chemical sintering can be employed to eliminate electrically non-conductive organic additives and surfactants that contribute to the increased resistivity in the electronics.[23] Among the thermal post-processing, laser sintering can conveniently be embedded in the printing process to prevent the warping of 3D-printed flexible devices.[10]

To demonstrate the capability of flexible electronics with external stimuli response, a triangular strain cycle can be applied to the strain sensor via a universal testing machine.[48] The strain sensor is connected to a digital multimeter to monitor the changes in its resistance. The same approach can be done by monitoring the voltage drop of the pressure sensor.[49] Other than flexible electronics' strain and pressure sensing characteristics, other real-time applications such as pulse monitoring, liquid evaporation monitoring, Morse code, etc. are also becoming popular.[47]

2.4. Hybrid technologies and systems

To achieve certain characteristics specific to electronic applications, multiple or hybrid manufacturing may be necessary to accommodate such needs. For example, Saari et al. used fiber encapsulation additive manufacturing (FEAM) and combined it with thermoplastic elastomer additive manufacturing (TEAM) to produce a composite capacitive force sensor.[43] FEAM and TEAM are both AM processes that may be utilized to fabricate integrated electrical components and soft thermoplastic elastomers, respectively. The researchers produced a sensor with a 3D-printed rigid frame, and wires with spiral patterns are embedded on the said frame. The sensor also has a dielectric spacer made of a thermoplastic elastomer that compresses upon applying force. They investigated the capacitance change versus the applied uniaxial compression load to evaluate the effectiveness of the capacitive force sensor.

Wehner et al.[44] used molding and soft lithography to fabricate the body and the microfluidic logic of a robot, respectively. In contrast, they used a multi-material embedded 3D printing method (EMB3D) to produce or pattern the other parts needed for movements, such as the catalytic reaction chambers and onboard fuel reservoirs. They concluded that these combinations of materials and manufacturing technologies could be applied in soft, autonomous robots. Table I summarizes 3D printing technologies and materials to manufacture flexible electronic devices for specific applications.

2.5. 4D Printing

Shi et al.[50] defined 4D printing as "the change of shape, property, and functionality from a 3D-printed structure response to time or external stimulus."[51] These stimuli include water, heat, light, and pH.[50] With the abovementioned capabilities, 4D-printed parts have been popular for soft electronics, soft tactile sensors, actuators, etc.

Zarek et al. fabricated shape memory objects intended for responsive and flexible electrical circuits through 3D printing of methacrylated monomers. These responsive objects may be used for sensors, soft robotics, minimally invasive medical devices, and wearable electronics.[52] Advincula and his team have recently published the 4D printing of an epoxy/polybenzoxazine copolymer and CNT composite that exhibited high thermal stability and shape memory printing.[53] Wu et al.[54] designed a 4D-printed flexible integrated magnetoelectric tactile sensor. The tactile sensor demonstrated a piezoelectric property (i.e. converting the external pressure into electric energy) even without any piezoelectric parts. The sensor consists of two parts. The first part is a porous structure fabricated by SLS using TPU/NdFeB composite powders. The second part is a helix structure with two flat plates fabricated by Selective Laser Melting (SLM) using 316 stainless steel powders. The device assembly can transfer mechanical to electrical energy (and is based on the electromagnetic introduction principle). With the self-powered, sensitive, and quick-responding properties, this 4D-printed tactile sensor may be used as a pressure sensor applicable to warn against illegal invasion. This study also provides a new concept for the functionality-changed and property-changed 4D printing.

2.6. Sustainability aspects

An electronic circuit could be found in many devices we use daily. It would be essential to manufacture these devices and electronic components considering aspects related to sustainability (e.g. green energy, production efficiency, green environment, life cycle assessment, industry 4.0, etc.). The traditional method of manufacturing electronic circuits involves multiple production steps and consumes much energy aside from using toxic/corrosive etching chemicals to remove unwanted photoresist materials. On the other hand, novel manufacturing by digital printing reduces the manufacturing steps, energy consumption and waste production. It does not require etching, as only the needed material is selectively deposited where it is required. Printing electronics is a disruptive innovation method that manufactures electronic components used for many devices, including solar panels, energy harvesters, photovoltaic cells, and various sensors. Printing of electronics requires a transdisciplinary research and production environment of different fields such as materials engineering, electronics architecture, robotics, automation, Artificial Intelligence, and process optimization.[55)]

One example presented by Kunnari et al.[56] is the printing of a wristband. The researchers assessed the environmental impacts of inkjet printing (using a preliminary screening approach) and compared it with the traditional process using a printed wiring board. They also evaluated the inkjet-printed interconnections and compared them with the other parts of a prototype product. They observed that it is possible to make electronics printing consume less energy than the traditional method. However, they claimed that more information is needed (e.g. process-specific data for both the materials and printing processes), specifically, the manufacturing data for the ink and nanoparticle. Factors such as printing speed, substrate cleaning, and yield should all be considered for the printing itself. Optimization of all the involved processes is needed before claiming that the printing process is environmentally friendly. Another involves the possibility of using biobased additives or feedstocks as in the use of nanocellulose and cellulose-based composites[57,58] or hydrogel inks[59] for sustainable 3D printing materials.

In the next section, the authors elaborate on the biggest challenges we face in advancing the manufacturing of flexible electronics and how the future might look as we address some of these challenges and take on research and development advancements.

3. Challenges and prospects

Flexible and printed electronics carry opportunities and challenges for new materials and function development. The actual development of integrated devices has been possible. Still, shape conformality, high performance, and operating environmental considerations will always necessitate optimized research and development protocols for high technology readiness level requirements.[1,55] There is also the need to have a hierarchy of fabrication and assembly methods that will require multi-materials approaches from metals, semiconductor films, conductive inks, and nanocomposite active and passive components.[4,10] The use of elastomers for flexible shape conformality should be married with the environment and chemical resistance against degradation of performance.[42,49,52] The materials' processing conditions and intrinsic development of structure-composition-property will determine the structural hierarchy from nano- to micro- and macroscopic structures. Thermo-mechanical properties based on tensile, compression, and flexural modulus will be integrated with the semiconductor and resistance properties of the conducting elements and will have defined actuation movement.[10] These are the possible requirements for unique wearable and soft-robotics types of applications.

Here is an overview of the prospects on research trajectories for flexible electronics, as summarized in Fig. 2. The figure shows that there can be two major directions that will continually inform each other: the tunability of characteristics of flexible electronic devices and the innovative manufacturing methods.

3.1. Flexural properties and sustainable materials

New thermoplastic and thermoset elastomer composites that take advantage of sustainable materials, e.g. biobased materials, natural fibers, that take advantage of the cost and sustainability engineering with proper life-cycle analysis and

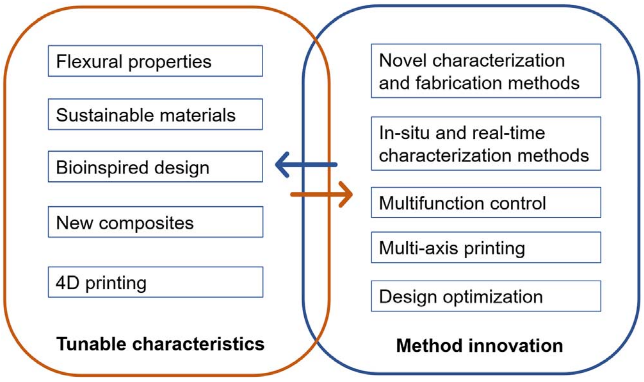

Fig. 2. (Color online) Potential research directions for flexible electronics.

overall techno-economic analysis (TEA) of renewable or recycled materials in conjunction with the AM methods.

3.2. Multifunction control and a new breed of composites

Multifunctional utility, i.e. combined optical and acoustic sensing, electro-optical modulation, wireless multi-frequency communication, etc. This should be paired with appropriate linear versus non-linear thermo-mechanical behavior as a flexible substrate with both synthetic and bio-based polymers sources. The use of fiber composites, e.g. C-fiber, Kevlar, PP fibers, natural fibers, etc. as integral strengthening materials in elastomers will be explored.

3.3. Bioinspired design and 4D printing

3D printing with bioinspired design and function that draws inspiration towards various applications and new structure-composition-property relationships.[32] For example, stimuli response, e.g. motivation from metamorphic bone and exoskeleton structures, color-or shape-changing in marine organisms, and controlled wetting behavior in insects, plants, and animals. All could lead to new wearable device designs and electronic functions, which may lean more towards the direction of 4D printing.[60]

3.4. Development of novel characterization methods

This will look at the use of high resolution and hierarchical imaging and chemical spectroscopy, as well as with signal transduction or logic circuit operation. They will determine meta- and trans-formative behaviors with appropriate stimuli that can be quantified with methods like localized surface plasmon resonance (SPR), surface acoustic wave (SAW), and piezoelectric effect with spectroscopic probe measurements (IR, Raman, Fluorescence, etc.).

3.5. Novel fabrication and in situ and real-time characterization of CNC and multi-axis printing

This will take advantage of the anisotropic ordering of the electrical or optical properties and the ability to control their bias with direction. This can be facilitated by a combination of reactive curing or ordering via shear forces and extrusion in FDM and DIW printing methods or by light orientation in optimized dispersions for photo-induced stereolithography or digital light processing (SLA/DLP) and binder jet fabrication methods.[37,48]

3.6. Optimized feedback loop in design

Building a database and algorithmic artificial intelligence and machine learning methods for optimizing device fabrication from multi-material and fabrication orientation methods.[61] This will enable the orientation of property control and high-performance conductivity, thermo-mechanical properties, and actuation performance in specific operating environments.[62]

4. Conclusion

This review aims at contributing to the body of knowledge on flexible electronics by providing a brief overview of its recent developments, methods and technologies for manufacturing, materials processing, post-processing and testing, state-of-the-art applications, the major challenges, and the future of research in this area. Previous and current researchers have come to a consensus that the biggest challenge lies in sustaining, tuning, and improving its flexural characteristics. In addition, further research and interdisciplinary inquiry are needed to explore novel characterization and fabrication techniques, optimized design processes, and formulation and functionalization of materials with tunable properties, and sustainability aspects that necessitate pushing for innovation and that would cater to the evolving demands for specific applications from the industry, and the fields of biomedicine, sciences, and engineering.

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