4D Printing of Electroactive Materials

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In recent years, the intersection of 3D printing and “smart” stimuli-responsive materials has led to the development of 4D printing, an emerging field that is a subset of current additive manufacturing research. By integrating existing printing processes with novel materials, 4D printing enables the direct fabrication of sensors, controllable structures, and other functional devices. Compared to traditional manufacturing processes for smart materials, 4D printing permits a high degree of design freedom and flexibility in terms of printable geometry. An important branch of 4D printing concerns electroactive materials, which form the backbone of printable devices with practical applications throughout biology, engineering, and chemistry. Herein, the recent progress in the 4D printing of electroactive materials using several widely studied printing processes is reviewed. In particular, constituent materials and mechanisms for their preparation and printing are discussed, and functional electroactive devices fabricated using 4D printing are highlighted. Current challenges are also described and some of the many data-driven opportunities for advancement in this promising field are presented.

1. Introduction

3D printing is an additive manufacturing (AM) process that has gained widespread popularity in recent years as an accessible, low-cost method of rapid prototyping and fabrication. 3D printing has applications in medicine, food processing, robotics, and materials research, where it has been used to fabricate complex metamaterials for specialized applications.[1] As a form of rapid prototyping, modern printing processes can shorten the time to market for product development, reducing costs and complexity.[2] This is a consequence of the ability of 3D printing to provide a direct means for fabrication, replacing traditional multistep processing sequences.[3] Furthermore, 3D printing processes are capable of building complex geometries at high resolution without the use of specialized tooling, which can be useful in a wide variety of applications.[4,5]

Several distinct 3D printing processes exist, but all are based on AM on a layer-by-layer basis. Prior to printing, a 3D computer-aided design (CAD) model is generated and “sliced” into a series of layers, and the processing parameters for each layer are written to a file in the form of code. The printer accepts this code, allowing it to build up the object represented by the original model. The most common types of 3D printing include direct ink writing (DIW), whereby liquid ink is deposited selectively and cured by exposure to light or another curing agent; fused filament fabrication (FFF), whereby a thermoplastic filament is extruded through a heated nozzle and allowed to solidify; stereolithography (SLA) and digital light projection (DLP), whereby a layer of liquid photopolymer resin is selectively cured by exposure to UV light; and selective laser sintering (SLS), whereby a powder material is sintered and fused together, forming a solid material. Hence, the printing process is strongly dependent on the limitations of both the software (CAD programs and slicer programs) and hardware (such as laser spot size and nozzle diameter).

Recent advances in both software and hardware have allowed for the fabrication of unique structures and materials; for example, multimaterial processes have enabled the direct printing of composites with highly tunable properties in a single step.[6]

The rapid expansion of 3D printing processes has led to higher resolutions, improved print quality, and a wider selection of printable materials. In recent years, the ability to directly print functional devices has emerged as an important subset of 3D printing, especially by the use of novel materials with functional characteristics.[7] The concept of 4D printing, attributed to Tibbits,[8] extends this idea to the intersection of 3D printing and functional materials that are capable of responding to an external stimulus.[9]

Specifically, the “fourth dimension” of time represents a predictable, characterizable stimulus response that provides a backbone for the functionality of a printed device. When exposed to a stimulus such as heat, moisture, light, or electric current, structures undergo a change in material properties or shape[10] over a length of time. These characteristics may be tuned and “programmed” into the specimen. 4D printing has been used to create self-deploying structures, actuators, hinges, grippers, and personalized medical devices.[11]

Specifically, electroactive printable materials present a promising direction within the emerging field of 4D printing (Figure 1). In recent years, the development of printable electroactive inks for film-based applications has demonstrated the high demand for faster, inexpensive, and direct methods of creating electroactive devices.[12] These materials have applications in...
memory storage, RFIDs, and artificial muscles for robotics. The advent of 4D printing expands this reach to fully printable structures that do not require a film or other substrate. Hence, printable electroactive materials represent a "print in place" technique, whereby whole functional parts are created entirely from start to finish on a single device. With 4D printing, the landscape of electronic devices, actuators, and robotics has the potential to reap the time-, cost-, and material-saving benefits of AM.

As research on both printing processes and printable materials develops, so too will the field of electroactive 4D printing. In this Review, an overview of current electroactive materials and the mechanisms by which they are fabricated and utilized is presented. Because the nature of the printing process strongly dictates the material characteristics and properties, the sections are grouped by print process.

## 2. Ink-Based 4D Printing of Conductive Materials

### 2.1. Process Overview

DIW is an AM technique that utilizes a computer-controlled stage and an ink-dispensing nozzle to generate 3D geometries and designs. DIW processes can be divided into filament-based approaches and droplet-based approaches. In either case, the basic fabrication mechanism involves pressurizing the nozzle for ink extrusion. As it is extruded, the ink is directly deposited on a desired surface along a programmed path to build a specified structure in a layer-by-layer fashion. For proper ink flow from the nozzle and good shape retention of the structure upon deposition, the rheological and viscoelastic properties of the ink are crucial. Specifically, the ink must have a low enough viscosity to allow for sufficient ink flow upon pressurization of the nozzle, and after extrusion, the material must undergo a rapid increase in stiffness to maintain its shape.

Benefits of DIW include the ability to print with a wide variety of materials, and the freedom to design and control the composition of the extruding ink, rather than being limited to proprietary materials produced for company-specific equipment. In addition, the setup requirements for a DIW system are relatively simple and inexpensive, and the printing process itself is highly controllable, with the barrel volume and nozzle size easily changed. A high level of resolution of printed parts can be achieved through the use of fine micronozzles.

### 2.2. Materials and Mechanisms

Several different active materials have been used successfully to produce ohmically responsive 4D printed structures via DIW. Colloidal nanoparticle (NP) inks made with metallic powders are commonly used as conductive inks in DIW, and can be used on their own or combined with other smart material elements for electronically stimulated activation of 4D printed devices.

![Figure 1](image-url) An overview of the 4D printing of electroactive materials as a subset of 4D printing, which is the intersection of 3D printing and “smart” materials. This article characterizes the current state of research in the 4D printing of electroactive materials by print process type, with further discussion on materials, mechanisms, and printable devices.
Figure 2. 4D printed structures fabricated with DIW techniques. A) Illustration of direct print setup for (top) filament-based and (bottom) droplet-based approaches. Adapted with permission.\(^{[21]}\) Copyright 2004, Elsevier. B) Chitosan actuator bending under an applied voltage. Adapted with permission.\(^{[33]}\) Copyright 2018, MDPI. C) Electroactive knit textile made of twined 3D printed LCE/Agsis conductive fibers that can be activated to form a) a dome and b) a blooming-like structure. Adapted with permission.\(^{[44]}\) Copyright 2019, American Chemical Society. D) 4D printed sequentially folding box, activated using resistive Joule heating of LCE strips. Adapted with permission.\(^{[26]}\) Copyright 2017, Royal Society of Chemistry. E) 4D printed SMP electroactive smart gripper. Adapted with permission.\(^{[42]}\) Copyright 2019, American Chemical Society. F) Demonstration of the shape-changing capabilities of 3D printed electroactive liquid-detection SMP sensor. Adapted with permission.\(^{[43]}\) Copyright 2019, Elsevier.
postprinting curing processes, using either heat or UV light may be required.

Liquid crystal elastomers (LCEs) are a type of soft active material that can undergo rapid, significant, and reversible changes in response to various external stimuli, including heat, light, and electronic and magnetic fields. Actuation of LCEs is due to a temperature-dependent transition from the nematic state to the isotropic state, which occurs at the isotropic transition temperature \( T_{NI} \). Much work has been done by various groups to develop an LCE ink that can be printed and prepared using DIW printing, mainly by combining LCE monomers with photoinitiators and amine chain extenders to print the LCE, normally in its nematic state. However, to obtain proper ink viscosity, elevated printing temperatures were required, which resulted in unfavorably high \( T_{NI} \). Notably, Zhang et al. managed to develop a formulation that removed these additives with a single-component biphenyl-based liquid crystal ink that could print the LCE while in its isotropic state. Although the ink \( T_{NI} \) was at a more reasonable temperature range (50–65 °C), the required printing temperature remained quite high (200 °C). DIW offers a convenient way to prepare LCEs as the shear forces from ink extrusion can be used to order the liquid crystal, which can then be set with in situ curing. In addition, on their own, LCEs can only achieve unidirectional responses when cycling between nematic and isotropic states. For more interesting shape changes, a multimaterial scheme is necessary, which makes LCEs prime candidates for use in 4D printed structures.

Polyelectrolyte hydrogels are soft, responsive materials that experience recoverable deformation upon exposure to an electric field. The hydrogel response varies with the gel’s polyion concentration, which can be used to control whether the gel shrinks (low polyion concentration) or swells (high polyion concentration) in response to a potential gradient. Deformation is also dictated by the osmotic pressure differential that occurs from an ion concentration difference developed between the inside and outside of the hydrogel upon application of the electric field. Upon removal or polarity change of the applied electric field, shape recovery can be achieved. Polyelectrolyte hydrogel inks are generally prepared by mixing a polyionic powder, typically chitosan or gelatin, into an acidic solution for up to several hours to ensure proper homogeneity before sonication or vacuum drying. The ink is then removed. Application of different solutions, such as ethanolic sodium hydroxide (EtOH-NaOH), between deposited layers during the printing process may also be utilized to ensure rapid solidification of the printed ink.

Shape memory polymers (SMPs) are a class of active materials that can recover their original shape from potentially several deformed states in response to external stimuli. Thermostressive SMPs are the most broadly studied for the ease of activating the shape memory effect (SME) and ability to tune parameters such as the transition temperature. Typically, an original, or permanent, state is fixed by deforming the polymer to a specific strain while it is above some transition temperature, generally its glass transition temperature \( T_g \) or melting temperature \( T_m \). Partial deformation states are achieved by cooling the material at a constant stress before unloading the system. Upon reheating the polymer above its transition temperature, the material returns to the programmed original state. SMPs filled with conductive fillers such as carbon nanotubes (CNTs), carbon nanofibers (CNFs), or carbon black (CB) allow for electroactivation of the SME. The ability to use electricity for internal resistive Joule heating to trigger shape change in SMPs is attractive for several reasons, including hands-free activation, more uniform heating, and improved thermal conductivity for more rapid material response in comparison to direct external heating. These conductive SMPs fall under the class of electronic functional materials known as conductive “polymer nanocomposites” (CPNs), which have promising applications in various types of sensors and other printed electronics.

2.3. Devices and Applications

2.3.1. Temperature-Sensitive Actuators

A number of temperature-sensitive actuators stimulated through Joule heating have been made through the DIW process. Yuan et al. combined prefabricated LCE strips \( T_{NI} \approx 87 \degree C \) with 3D printed elements, including a multimaterial ink-jetted soft matrix and DIW silver ink for conductive wires. Actuation of the LCE strip was done via Joule heating from the printed wires, controlled by an external power source. When current was supplied, the silver wires conducted heat to the LCE, which contracted in length, causing the structure to bend. When the current was turned off, the temperature gradually dropped below the \( T_{NI} \), and the hinge regained its undeformed shape. By varying the current value supplied to the hinge, the authors were able to characterize the bending behavior, which saw an increased bending angle and rate with an increased current value above a determined threshold. From these experiments, they were further able to design and create similarly actuated active structures, including a soft airplane, miura-origami structure, self-folding box, and locomotive soft crawler. Figure 2B shows the full folding and unfolding sequence of the 4D printed box.

Subsequent efforts focused on incorporating DIW-printed LCEs with lower \( T_{NI} \) for increased practicality of 4D printed temperature-sensitive designs. Roach et al. developed an LCE ink that could be printed via DIW at ambient temperature to use as a soft, reversible actuator in their 4D printed structures. By incorporating the thiol 2,2’-(ethylenedioxy)diethanethiol (EDDET) with the mesogen RM257 in a 1:1.15 molar ratio, they were able to achieve an LCE with a \( T_{NI} \) of about 42 °C. They developed a multimaterial 4D printed hinge with their novel LCE using Joule heating actuation through DIW-printed wires and then expanded on this design to create several more complex 4D structures, including a sequentially folding box, robotic gripper, and a soft robotic hand capable of signing American sign language letters. Roach et al. later explored potential smart-textile applications by developing a method to print LCE fibers using DIW. In a three-step fabrication process, LCE fibers were created by first extruding the ink onto a rotating mandrel, convectively heating the fibers for partial curing, then stretching the fibers between two mandrels for mesogen alignment. By weaving the LCE fibers together with Agsis conductive fibers, textiles capable of sequential folding from Joule heating activation were fabricated. Using Joule heating from a 2 A current, both an
expanding dome and a flower-like blooming structure were observed from the same textile (Figure 2C).\[44\]

### 2.3.2. Electroresponsive Devices

Various electroresponsive devices have been fabricated using DIW. Zolfagharian et al. developed a Takagi–Sugeno (T–S)-type fuzzy model and a discrete rigid finite element model to predict the bending behavior of 3D printed porous polyelectrolyte hydrogel actuators in response to a certain applied voltage. They verified their models by printing a polyelectrolyte hydrogel actuator using a chitosan and acetic-acid-based ink and measuring the end-point deflection of the actuator in response to various applied voltages while submerged in an electrolyte solution. Both models were found to match exceedingly well with the experimental results.\[31\] Zolfagharian et al. went on to study how the type and concentration of electrolyte solution the actuator is submerged in affect the actuator’s bending behavior. They 3D printed chitosan actuators with arbitrary porous sizes and patterns and studied their bending behavior in two different electrolyte solutions, NaOH and NaCl, over six different concentrations varying between 0.1 and 0.2 M under a constant applied voltage of 5 V. It was found that actuators achieved higher deflections in the NaOH solution, and that for both solutions, an optimal molar concentration of about 0.12 M resulted in the maximum bending angle. The bending behavior of the actuator under an applied current can be seen in Figure 2D.\[33\] Zolfagharian et al. then used a stiffness-based topological optimization to develop a porous chitosan actuator design with maximal bending deformation. When comparing topologically optimized actuators with actuators with uniform lattices, they found the topologically optimized design had the greater end-point deflection.\[35\]

### 2.3.3. Shape Memory Structures

Several novel applications of DIW 4D printed shape memory structures have been used. Rodríguez et al. created electrically conductive DIW-printed functional structures from an SMP composite made of a CNF-filled epoxidized soybean oil (ESBO) and bisphenol F diglycidyl ether (BFDGE) resin. Composites with a CNF volume fraction of 5.6% had an electrical conductivity of 0.4 S cm\(^{-1}\), allowing for electrical stimulation of the SME. After programing the part to set its deformed shape, the printed part ends were connected to a 20 V, 150 mA power source to complete the circuit and allow for resistive heating of the composite. As it reached temperatures above its \(T_p\), the composite part gradually changed shape, with complete shape recovery after 180 s.\[39\] Combining the conductivities of metallic and carbon-based fillers, Wei et al. produced 4D printed electroactive shape memory structures with incredible conductivity and not requiring postprocessing curing treatments through a novel ink formulation of hybrid silver-coated CNFs (Ag@CNFs) in a PLA matrix with a dichloromethane (DCM) binder. The ink could be deposited via DIW at ambient temperature to produce self-supporting structures with conductivities greater than those of all carbon-filler-based CPNs, pure CNTs, and graphene. Using a 10.6 vol% Ag@CNF/PLA ink, a 4D printed smart gripper capable of grabbing a 5 g bolt when connected to a 1 V, 0.1 A power source and resistively Joule heated was fabricated (Figure 2E). It was able to grab the bolt in 20 s, demonstrating the printed NPC’s potential use for fast, low-voltage actuation.\[42\] Wan et al. fabricated electroactive shape-changing sensors by combining the shape memory capabilities of crosslinkingpoly(D,L-lactide-co-trimethylene carbonate) (c-PLMC) with CNT fillers. A 10 wt% CNT ink formulation was used to print conductive scaffolds that were then immersed in various solvents to measure the relative resistance change of the NPC due to swelling of the polymer matrix. As the polymer swells, the distance between the CNT fillers increases, causing a change in the electrical conductivity, thus allowing for liquid detection. As shown in Figure 2F, by applying an electric field, they were able to deform the sensor to adapt to different liquid levels while also maintaining a similar sensitivity to the solvent, demonstrating the possibility of 4D printed environment-adapting liquid sensors optimized for different solvent conditions.\[43\]

### 3. Filament Deposition 4D Printing of Conductive Materials

#### 3.1. Process Overview

FFF is an extrusion-based additive manufacturing process that utilizes the reversibles, temperature-dependent properties of a thermoplastic material. FFF is among the most common 3D printing technologies currently available, and many organizations have developed low-cost desktop printers that are able to produce parts using this process. The use of FFF has also begun to increasingly replace traditional manufacturing processes for rapid prototyping.\[45\] In recent years, FDM has demonstrated significant growth in the field of materials research.\[46\]

In an FFF process, a continuous thermoplastic filament (such as polyactic acid, PLA, or acrylonitrile butadiene styrene, ABS) is heated to a melting temperature and extruded using rollers or another form of applied pressure. The filament is then deposited on the build platform in a raster pattern; when the extruded thermoplastic cools on the build platform or on the previous layer, it hardens again, forming a solid part. After each layer is complete, the build platform or the extruder moves by a vertical distance equal to the height of a layer, and the process repeats again. Like all AM processes, the quality and success of FFF-produced parts are highly dependent on the print process parameters, including extrusion head temperature and speed, infill density, and layer height.\[47\]

#### 3.2. Materials

Using the FFF process in 4D printing often involves first producing a conductive or otherwise electroactive filament compatible with the extrusion process. FFF-printable conductive materials used to create 4D printed parts have been fabricated most often using a composite of two materials: a conductive, often carbon-based filler and a thermoplastic matrix, often PLA.\[48\] Several commercially available conductive filaments consist of a carbon–PLA composite. Common conductive fillers include multiwalled CNTs,\[49\] graphene,\[5\] carbon nanoparticles,\[50\] and/or carbon black.\[51\] To create a printable filament, these fillers
are commonly introduced into the matrix material prior to printing by means of a twin-screw extruder, [52] melt extrusion, [53] or a drawing process. A chemical solvent may also be used to disperse the conductive particles in the matrix material; the solvent is then evaporated from the solution. Dispersion into the matrix may be homogeneous or selective; a “local enrichment strategy” was developed by Shi et al. to deposit CNTs onto a printable PLA filament in favorable surface locations (Figure 3A). Using the local enrichment strategy, printed parts demonstrated an increase in electrical conductivity of eight orders of magnitude over CNT/PLA composites manufactured using other dispersion methods. [54]

Other matrix materials used to create conductive filaments include ABS, [55] poly(methyl methacrylate) (PMMA), [56] and a biodegradable polyester. [57] Dorigato et al. used a melt compounding process in conjunction with compression molding to create a printable ABS/CNT filament. Both the electrical and thermal conductivity of the ABS were strongly influenced by the weight fraction of CNTs added. [58] Grinberg et al. ultrasonically dispersed barium titanate, a piezoelectric material, into a matrix of polyaniline (PA) 11. The solution was formed into particles, then melted and extruded into filament form (Figure 3B). The filament was then used with a commercial FFF 3D printer having standard nozzle dimensions. [59]

3.3. Devices and Applications

3.3.1. Printable Sensors

Chemical sensing devices have been fabricated using direct FFF printing of conductive materials. In general, in the presence of an external stimulus, these devices produce a measurable change in one or more electrical properties (e.g., capacitance or resistance). Abdallah et al. demonstrated a capacitive fluid level sensor printed with a PLA/graphite filament. The sensor consists of two interdigitated printed parts and is able to detect liquid levels of up to 3 mL based on changes in the system capacitance (Figure 3C). An amount of fluid present between the printed capacitor “plates” causes a change in the dielectric constant of the two-phase medium between the plates, changing its capacitance. Results showed a strong correlation between the measured capacitance and the liquid level within the sensor’s digits. [60]

A resistance-based chemical sensor was printed by Aliheidari et al. from a composite of thermoplastic urethane (TPU) and CNTs. When the sensor was immersed in ethanol, it was found that the resistivity of the printed part changed over time; adjusting the infill density, CNT content, and extrusion orientation all affected the sensitivity of the device to ethanol. [61] Such a device has potential to be used for chemical leakage detection. Shemelya et al. used wires embedded within an FFF-printed part to...
fabricate a printable capacitive sensor. A print–pause–print approach was used to temporarily interrupt the FFF process when a copper wire or mesh was inserted. The resulting device was used to distinguish between three metal samples (nickel, aluminum, and copper) and two water samples (distilled water and salt water) by means of a change in capacitance.

In addition to fluid and chemical sensing applications, printable conductive materials have been used to fabricate stress and strain sensors. Matrix or substrate materials used in these sensors exhibit a high degree of mechanical strain recovery, so they are functionally reusable with minimal drift. Leigh et al. produced a printable conductive composite filament compatible with off-the-shelf FFF printers by adding carbon black at 15 wt% in a polycaprolactone (PCL) matrix. This material demonstrated piezoresistive characteristics and was successfully used as a sensor as well as a capacitive interface device capable of detecting finger-point touches. A fully printable multiaxial force sensor (Figure 3D) has been developed by Kim et al. using TPU and a TPU/CNT composite. The printed device exhibits a detectable change in resistance when a force is applied, and is sensitive to the submillimeter scale in three directions. Melnykowycz et al. fabricated a monofilament based on carbon black particles dispersed in a matrix of styrene–ethylene/butylene–styrene (SEBS) and demonstrated a measurable change in resistance of the printed filament when a strain was applied.

3.3.2. Functional Actuators and Devices

Several postprocessing techniques also exist to create a functional part using a conductive element and a printable element, although they do not utilize direct printing of conductive materials. Shao et al. developed a printable, electrically controllable gripping device by introducing silver nanowires into grooves within a printed part (Figure 3E). When the silver-based solution dried, the resulting composite became electroactive due to the presence of a conductive pathway. Consequently, the deformation of the gripper could be precisely controlled by varying the supplied voltage.

Carro et al. used a chemical postprocessing step to hydrolyze a printed ionic polymer–metal composite (IPMC). A printable “precursor” composite of Nafion and a thermoplastic filament was used to manufacture a specimen using FFF. Then, an electrochemical plating process was used to create electrodes at the surface of the printed part, rendering the part functionally active. Using this material, the authors printed an IPMC actuator that exceeded the response performance of a conventionally-manufactured IMPC actuator at the same input frequency. However, the response and relaxation times of the printed actuator were both slower compared to the conventional part.

Using a combination of “fiber encapsulation additive manufacturing” (FEAM) and “thermoplastic elastomer additive manufacturing” (TEAM), Saari et al. demonstrated a printed capacitive touch sensor. The electrodes of the capacitor were printed using the FEAM process, which allows a conductive wire to be fully encapsulated within a matrix material extruded by a nozzle. Here, the matrix is a rigid, ABS-based material. TEAM was used to fabricate the dielectric component of the capacitor from a compliant thermoplastic elastomer. The three-layered sensor was created monolithically using a single custom-built 3D printer. Characterization of the device showed a strong correlation between the applied load and measurable capacitance; however, a minimum loading is required before a capacitance response occurs. The authors attribute this fact to the surface roughness of the last printed layer, an artifact of the FFF-based process used to build the sensor. However, the use of FEAM and TEAM shows promise due to the variety of elastomeric materials compatible with the process, allowing the sensitivity of the device to be easily tuned.

3.4. Shape Memory Applications

Recently, the development of directly printable SMPs has enabled the 4D printing of active devices that can reveal the presence of one or more stimuli. The advent of 4D printable SMPs using FFF greatly simplifies the fabrication of these materials, often to a single printing step.

Liu et al. used silicon carbide (SiC) and graphite powder in a PLA matrix to fabricate a conductive-printed SMP. The resulting part exhibits reversible temperature-activated behavior after a shape is initially “programmed” at 90 °C. Testing showed that there was a strong correlation between the measurable conductivity of the material and the shape recovery time, and the composite exhibited up to a tenfold decrease in shape recovery time compared to pure PLA. The use of polycyclooctene (PCO), an excellent SMP candidate material with recoverable strain of over 700%, as the base of a printable filament was demonstrated for the first time by Liu et al. Boron nitride (BN) and CNTs were added to the PCO matrix to form a multi-responsible conductive composite. After printing, gamma-ray irradiation was used to crosslink the functional filler materials within the printed part. The crosslinked material exhibited a high degree of shape recovery under electric current and under illumination.

Micalizzi et al. produced a set of multimaterial SMP actuators using dual-extrusion FFF printing. Within each actuator, a set of “active blocks” printed using a carbon-based conductive filament was embedded in a base printed using pure PLA. The active blocks functioned as local heating zones, allowing for controllable deformation in a predictable manner. The actuators demonstrated a temperature-based activation and recovery cycle. Zeng et al. also used a dual-extrusion process to print a continuous carbon-fiber-reinforced PLA composite. The composite part was activated using electric resistance heating and exhibited a shape recovery rate of over 95% (Figure 3F). Compared to traditionally printed materials, the carbon fiber–PLA composite exhibits a marked improvement in mechanical strength.

4. Light-Processing 4D Printing of Conductive Materials

4.1. Process Overview

Digital light processing (DLP) is a type of vat photopolymerization AM technique. By coupling a light source with a photoreactive polymer, a complex 3D structure can easily be fabricated via DLP at low cost. Other advantages of the DLP process include high dimensional accuracy, high resolution (50 μm minimum
that in The light exposure time, the wavelength of the light source, by the speci

![Image](www.advintellsyst.com)

[80] Fang et al. developed a stretchable and biocompatible in conductive DLP printing is carbon porous nanocookies at least 7 days after mixing. The other carbon nanomaterial used onstrated excellent dispersion, with no sedimentation evident for sile fracture stress of up to 33%. The composite resin also dem-

![Image](www.advancedsciencenews.com)

[74] smooth surface finish, isotropy in the printed part, and a wide range of material choices with varying properties, such as water resistance.

DLP printers often include a digital projector, a light source, a material tank, and a build platform. The build platform is dipped into the material tank containing the photoreactive polymer in the liquid state. The bottom of the tank is transparent, so the light pattern produced by the digital projector can pass through the tank, selectively solidifying specific regions of the material. After curing the first layer, the platform moves up by the specified layer height to cure the next layer of the part. The light exposure time, the wavelength of the light source, and the amount of power supplied are the three main parameters that influence the quality of the printed part.

SLA is another common type of vat photopolymerization AM technique. SLA and DLP have similar working principles; however, in the SLA process, the projector is replaced with one or more lasers, often in the UV range, that are free to move underneath the material tank. During the printing process, the lasers sweep over the region to be cured, selectively polymerizing the resin which is exposed to the beam. Compared to DLP, which can cure the entire layer with a single projection, the SLA process requires more time. Additionally, DLP often uses a conventional light source instead of one or more laser modules, making it more accessible than SLA.

4.2. Materials

Conventional 4D printing of conductive materials often requires high temperatures during the fabrication process (such as FFF). Therefore, advanced materials such as nanocomposites are a better match for DLP printing. As DLP prints under ambient conditions, the insulating portion and the conductive portion of a part can be produced simultaneously. This can make the fabrication process faster and reduce the risk of damage to low-temperature polymer insulation layers.

Carbon nanomaterials are one of the most popular conductive materials used in both DLP and SLA printing. Mu et al. used multiwalled carbon nanotubes (MWCNTs) mixed with an acrylic-based photocurable resin to 4D print conductive complex structures. MWCNTs of more than 93% purity were fabricated from a chemical vapor deposition process. After the bulk MWCNTs were dispersed using ultrasonication and ball mill techniques, they were mixed with the resin and ready to use. Sandoval et al. demonstrated the use of MWCNTs with a modified commercial SLA printer. By varying the concentration of MWCNTs in various resins, the authors demonstrated an increase in tensile strength of up to 7.5% and an increase in tensile fracture stress of up to 33%. The composite resin also demonstrated excellent dispersion, with no sedimentation evident for at least 7 days after mixing. The other carbon nanomaterial used in conductive DLP printing is carbon porous nanocookies (NCs). Fang et al. developed a stretchable and biocompatible conduit for regenerating peripheral nerves using a DLP printer (Figure 4A). The conduits are made of reduced graphene oxide (GO) nanosheets and NCs. The GO is synthesized using graphite powder, sulfuric acid solution, and sodium nitrate. Potassium manganate (VII) and hydrogen peroxide were then added to precipitate GO. Then GO was used as the raw material for synthetic NCs.

The other type of conductive nanomaterial used with DLP printing is silver nanoparticles. Unlike commonly used bulk silver, the silver nanoparticle used in DLP printing is made from a photoinduced redox reaction. It is a room temperature reaction using a conventional light source, which is a perfect match with DLP printing. Yang used a particle-free reactive silver ink coupled with DLP printing technology to produce conductive silver patterns (Figure 4B). The conductivity of this silver nanoparticle is 40% of the conductivity of bulk silver. The resulting fabrication process is facile and suitable for making complex conductive patterns and pathways.

4.3. Devices and Applications

4.3.1. Circuit Elements

Stretchable circuits and hollow conductive structures are two of the main applications of DLP-printed parts. Mu et al. fabricated a hollow capacitive sensor, electrically activated shape memory composites, and stretchable circuits all using MWCNTs and a DLP printer. The fully 4D printed hollow capacitive sensor was capable of sensing compressive strains or stresses. Due to the hollow structure, the capacitive sensor could detect the change in spacing between two printed plates. In addition, this printed sensor can also be used for measuring liquid levels; the capacitance of the sensor increases with the internal liquid level. The authors also demonstrated a DLP-printed MWCNT structure with SME induced by electrical resistance heating. The SME is a very important feature in soft robotics, flexible electronics, and medical devices. Finally, the authors fabricated a fully DLP-printed MWCNT spring-like circuit that can be stretched by 100% of its original length, with only a modest (13%) increase in resistance. Furthermore, the resistance of the circuit nearly recovered to its original value when returned to the relaxed condition.

Cortés et al. showed that a higher Young’s modulus and glass transition temperature of DLP-printed CNT composites can be achieved by additional postcuring of printed parts. The CNT nanocomposite studied showed a linear response of electrical resistance when a strain was applied, making it a suitable material for strain sensors. Postprocessing does not have a significant effect on the electrical properties of the printed parts, enabling mechanical properties to be tuned while preserving the electro-active response.

4.3.2. Biological Applications

Many biological processes can be induced by a high-frequency magnetic field. Fang et al. used DLP-printed porous GO NCs for growth factor release and cell stimulation to promote neuron cell differentiation. The use of conductive NCs permits electromagnetic stimulation of the cells and subsequent differentiation and proliferation of neuron cells in vitro and in vivo. This application has the potential of regenerating tissue for curing neuronal diseases. The printed conduits demonstrated by Fang et al. had the ability to carry large amounts of neuron growth factor.
Figure 4. 4D printed structures using DLP techniques. A) Bioapplication for growth factor release and cell stimulation: a) fabrication process and materials used. b) NC@C under MF treatment facilitated magnetoelectric conversion for growth factor release and cell stimulation to induce neuron cell differentiation. c) Image of a 3D printed NC@C with elastic properties. d) The elastic properties of NC@C showed stretchable properties. e) SEM image of cells adhered on NC@C. The roughness of the exposed NCs on the surface improved cell attachment. f) CLSM images of cells proliferating on NC@C with microchannels. DAPI-stained nuclei (red) and phalloidin-stained F-actin (green). Adapted with permission.© 2020, Nature Publishing Group. B) Schematics illustrating the working principle and procedure of room temperature Ag printing. A commercial projector projects exemplary “UCLA”-patterned light through a digital mask on a substrate immersed in the photoreactive silver ink. Silver particles are simultaneously produced and settle on the substrate. Followed by chemical annealing with a NaCl aqueous solution, the conductive Ag pattern of “UCLA” is generated on an exemplary PET substrate shown in the photo. Adapted with permission.© 2019, Wiley.
that can exhibit high efficiency and protein permeability. In addition, the NCs conduits plus high-frequency magnetic field treatment can significantly increase the in vivo release rate of protein and doxorubicin, used as a model drug. The use of NCs in this application can help to improve the myelin sheath layers and direct the axon orientation. Using the same treatment in vitro can help to regain the diameter of animal muscle fibers.\textsuperscript{81}

5. Laser-Sintering 4D Printing of Conductive Materials

5.1. Process Overview

SLS is a type of 3D printing technique that uses a high-powered laser to melt or fuse a powdered material.\textsuperscript{75} SLS printers are capable of printing various materials such as metals, ceramics, and polymers. This makes SLS an especially promising method for the 4D printing of conductive materials. Unlike other powder bed fusion techniques, the powder being melted can be mixed with metals and polymers. The laser melts the polymer during the printing process. Afterward, the polymer becomes a binder between metal particles, and a 3D part is fabricated layer by layer (Figure 5A).

An SLS printer generally includes a laser, a powder bed, a roller, and a material storage container. A leveling roller spreads a thin layer of powdered material around the powder bed. Then, the laser melts a specific region of the powder to fuse those particles in the region and create patterns. The powder bed lowers and more material is spread on top to prepare for the next layer. This process repeats on a layer-by-layer basis until the print is done.\textsuperscript{84}

There are multiple benefits of using SLS printers. Unlike traditional FDM printers, SLS allows users to print hollow structures without the need for support material. This reduces the print time and saves significant material. Also, the material powder can be recycled after each layer, reducing the costs associated with wasted material.\textsuperscript{75} The resolution of the print job can be guaranteed by the laser used in the process. However, there are several limitations of the SLS process. First, the high-power laser used in the printer is expensive as is the cost to maintain the printer. It is also a high-temperature melting process (especially when polymer materials are used), so the operating environment needs a qualified ventilation system to keep the user safe.

In addition to SLS, another light-based process termed flash lamp-based processing (or flash lamp-induced processing) has attracted recent attention in printing conductive materials. Shin et al. used irradiation from a flash lamp as the energy source to transform metal oxide nanoparticles into metal film through reduction and sintering. During the process, a large amount of heat is generated by the incident light, such that both chemical and physical interactions reinforce the adhesion between a metal oxide layer and an insulation layer. The metal oxide layer is located between a metal film and the insulation layer. Here, the use of flash lamp-based processing can eliminate the inherent atomic lattice mismatch between the metal/insulation interface, resulting in outstanding robustness of the metal electrode.\textsuperscript{85}

5.2. Materials

As SLS melts a polymer rather than metal powder during the process, composites are often used to 4D print conductive materials. A polymer matrix with conductive fillers is the most common type of SLS-printable conductive composite. In general, the higher is the loading of the conductive filler constituent, the higher is the electric conductivity of the printed part; however, the mechanical properties of the part degrade and the part subsequently becomes harder to machine.\textsuperscript{86} Graphite nanomaterials are among the most popular materials in SLS printing. A 65 vol% loading of natural graphene (NG) in an epoxy matrix can achieve an electrical conductivity of 38 000 S m\textsuperscript{-1}.\textsuperscript{87} Different material preparation methods can also affect the electrical properties of the part. Graphene nanoplates (GNPs) dispersed in a polyaniline (PA) matrix using liquid-phase absorption followed by deposition exhibited a higher electrical conductivity than a GNP/PA composite fabricated using melt blending.\textsuperscript{88}

Unlike the SLS process whereupon a polymeric material is melted, flash lamp-based processing utilizes a chemical reaction to produce the conductive layer. Park et al. demonstrated a copper conductor produced using the flash lamp-based process with exceptional conductivity and stretchability. The conductivity of the copper electrode reached 37 kS cm\textsuperscript{-1} and the device exhibits the capability of attaining a strain of 20% in all directions. The authors demonstrated the potential of electrodes created by flash lamp-based processing via a device that permitted wireless near-field communication (NFC) on human skin.\textsuperscript{89} Im et al. used flash lamp-based processing to grow multilayer graphene (MLG) via the decomposition and photolysis of acetylene gas. The high energy from the lamp induced a rapid heating and quenching process, which triggered the chemical reaction of the gases in the chamber, allowing for the synthesis of MLG directly on a nickel film.\textsuperscript{90}

5.3. Devices and Applications

5.3.1. Strain Sensors

Gan et al. demonstrated that SLS 3D printed TPU/single-walled carbon nanotube (SWCNT) composites (Figure 5B) are suitable for strain sensors due to their excellent performance in cyclic response. At a loading of 0.2 wt%, SWCNTs in a TPU matrix experienced no degradation in a time-dependent resistance measurement under cyclic mechanical bending loads. In addition, the piezoresistive behavior of this composite experienced no significant change through 1000 bending cycles.\textsuperscript{91} This performance not only shows that SLS 3D printed TPU/SWCNT composites are a stable material for use in strain sensors, but that this composite shows promise for integration with wearable electronics and human-computer interfaces.

5.3.2. Fuel Cells

An effective bipolar plate for a methanol fuel cell must have an electrical conductivity greater than 100 S cm\textsuperscript{-1} under set target parameters. Alayavalli et al. created methanol fuel cells using
graphite bipolar plates fabricated by SLS printers, which demonstrated a conductivity of over 200 S cm$^{-1}$, demonstrating the ability of SLS processes to directly fabricate components of fuel cells.$^{[92]}$

6. Challenges and Future Perspectives

By utilizing various AM methods, including FDM, DIW, and SLS, 4D printing technology has realized numerous novel applications using conductive material in medical fields, soft robotics, and electronic components. However, challenges still lie in the development of this technique. First, the printing efficiency and quality of 4D printing are relatively low compared to matured 3D printing methods. Common 3D printing anomalies such as delamination between the printing layer and discontinuity of the printing material can be amplified when encountering the multimaterial settings and complex printing structures used in 4D printing. Second, the functionality of 4D printing is constrained to very specific working conditions or by a certain number of operations. For instance, some widely used SMPs are not reversible in shape transformation and require extra programing...
into new shapes. Soft actuators based on 4D printed conductive circuits suffer from the limited lifespan of repetitive operations. Third, a trial-and-error process is usually applied to solve the inverse problem (i.e., unknown design for a target shape change) in the design of 4D printed structures. The creation of a functional structure largely depends on prior experience and is then verified by simulations or experiments. The lack of systematic methods can lead to inefficient design processes and suboptimal structures.

To tackle these challenges, numerous studies have been conducted to explore new materials and advanced methodologies. For example, advanced electroactive composites have been developed to realize complex bionic functions using carbon-black nanoparticles and PLA. By finely controlling the printing process and structure, a dual electric shape- and color-changing mechanism can be achieved.[93] Similarly, hybrid silver-coated CNFs (Ag@CNFs) have been designed to reach high electrical conductivity (i.e., $>2.1 \times 10^5$ S m$^{-1}$) and demonstrate quick electrical-response behavior.[94] These newly created composites largely expand the capability and durability of 4D printing applications. Beyond the advanced synthesis of novel materials, insightful methods such as machine learning algorithms can also be another powerful tool to aid in finding better materials and manufacturing processes. This can be attributed to the capability of such algorithms to accurately describe and predict patterns that make up the problem space.[95] Machine learning methods have been actively applied to related materials engineering and AM topics and have demonstrated promising progress.[96] It is believed that similar machine learning techniques can also be utilized in the 4D printing of conductive materials. In terms of the printing process, precise in situ anomaly detection has been performed in FDM 3D printing using deep neural networks.[97] Printing defects such as over and underextrusion are successfully located in real-time images. This camera-based anomaly detection system can be easily transplanted into other AM platforms, including DIW and SLS, to monitor the conditions of the 4D printing process. As the 4D printing process may involve deformation over time, the real-time print quality has an essential impact on the later product. Hence, with time-dependent input information, recurrent neural networks (RNNs) can be applied to recognize the tendency behind the stream of images.[98] Moreover, RNNs have been successfully implemented to realize the functionality of 4D printed conductive sensors.[99] The device is able to recognize dynamic epicentral human motions by detecting deformation from laser-induced crack structures and analyzing the temporal signal through RNN algorithms. Based on this fundamental work, extension studies can be conducted to interpret the deformation of the 4D printed structures in multiple functionalities by decoupling the signals using advanced machine learning methods. For example, a 4D printed skin sensor made of conductive circuits deforms under the effect of both thermal and stress changes. By evaluating the pattern of signals generated by different kinds of deformation, information such as temperature difference as well as the stress impulse can be obtained to detect body temperature or symptoms of strokes. Finally, machine learning methods have been demonstrated as an effective tool in designing active structures for 4D printing. An evolutionary machine learning algorithm has been applied to determine and optimize the distribution of passive and active material for target deformation.[100] For applications involving conductivity, the active material can be switched to a conductive material with added boundary conditions on continuity and modified target functions such as the desired electrical signal.

7. Conclusion

The field of 4D printable electroactive materials continues to evolve as a subset of 4D printing in general, itself characterized by the intersection of AM and “smart”, stimulus-responsive materials. We have examined the current state of 4D printable electroactive materials across the most common printing processes; thus far, electroactive materials compatible with DIW, FFF, SLA, and SLS have been used to produce a number of functional devices, sensors, and other responsive structures. The 4D printing of electroactive materials shows promise for self-actuating mechanisms in robotics, “smart” textiles, chemical and fluid sensors, printable circuits, and medical devices. The development of specialized printing and postprocessing techniques has demonstrated the ability of AM to surpass the challenges and limitations of traditional manufacturing methods for fabricating electroactive parts. Further, the synthesis of new metamaterials and printable composites, as well as the modification of widely available printable materials for electroactivity, allows for the direct fabrication of a vast array of materials with highly tunable and predictable properties, suitable for many applications. Another promising avenue is the integration of machine learning to the design and execution phases of the printing process; in addition, machine learning methods have the potential to refine and guide how printed functional parts behave and are controlled. These and other developments in AM will improve the efficiency of 4D printing processes and the accuracy of printable parts, expanding the boundary of what is achievable using this emerging technology.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

4D printing, additive manufacturing, electroactive materials, shape memory polymers, smart materials

[1] a) A. A. Giannopoulos, D. Mitsouras, S-J. Yoo, P. P. Liu, Y. S. Chatzizisis, F. J. Rybicki, Nat. Rev. Cardiol. 2016, 13, 701; b) C.-Y. Liaw, M. Guvendiren, Biofabrication 2017, 9, 024102;
c) K. Demir, Z. Zhang, J. Yang, G. X. Gu, Adv. Intell. Syst. 2020, 2.

d) N. Nachal, J. Moses, P. Karthik, C. Anandharamakrishnan, Food Eng. Rev. 2019, 11, 123.

e) G. X. Gu, C.-T. Chen, D. J. Richmond, M. J. Buehler, Mater. Horizons 2018, 5, 939.

f) J. Z. Gu, M. Sajid, M. M. Rehman, G. U. Siddiqui, I. Shah, K.-H. Kim, J.-W. Lee, K. H. Choi, Sci. Technol. Adv. Mater. 2018, 19, 243.

[2] E. Macdonald, R. Salas, D. Espalín, M. Perez, E. Aguilerà, D. Muse, R. B. Wicker, IEEE Access 2014, 2, 234.

[3] V. A. Lifton, G. Lifton, S. Simon, Rapid Prototyping J. 2014, 20, 403.

[4] a) C. Lindemann, T. Reiner, U. Jahner, R. Koch, Rapid Prototyping J. 2015, 21, 216; b) Z. Jin, Z. Zhang, G. X. Gu, Adv. Intell. Syst. 2020, 2, 1900130; c) B. C. Compton, J. A. Lewis, Adv. Mater. 2014, 26, 5930; d) G. X. Gu, M. Takafoki, M. J. Buehler, Adv. Mater. 2017, 29, 1700060.

[5] D. Zhang, B. Chi, B. Li, Z. Gao, Y. Du, J. Guo, J. Wei, Synth. Met. 2017, 217, 79.

[6] a) F. Li, N. P. Macdonald, R. M. Guitj, M. C. Breadmore, Anal. Chem. 2018, 91, 1758; b) L. R. Lopes, A. F. Silva, O. S. Carneiro, Addit. Manuf. 2018, 23, 45.

[7] J. Ruiz-Morales, A. Tarancón, J. Canales-Vázquez, J. Méndez-Ramos, L. Hernández-Afonso, P. Acosta-Mora, J. M. Rueda, R. Fernández-González, Energy Environ. Sci. 2017, 10, 846.

[8] a) M. Zeng, Y. Zhang, J. Mater. Chem. A 2019, 7, 23301; b) L. Wang, J. Liu, Sci. China Technol. Sci. 2014, 57, 2089.

[9] S. Tibbits, Archit. Des. 2018, 84, 116.

[10] a) F. Momeni, S. M. Mehd, N. Hassani, X. Liu, J. Ni, Mater. Des. 2017, 122, 42; b) X. Kuang, D. J. Roach, J. Wu, C. M. Hamel, Z. Ding, T. Wang, M. L. Dunn, H. J. Qi, Adv. Funct. Mater. 2019, 29, 1805290; c) A. S. Gladman, E. A. Matsumoto, R. G. Nuzzo, L. Mahadevan, J. A. Lewis, Nat. Mater. 2016, 15, 413.

[11] a) Z. Zhang, K. G. Demir, G. X. Gu, Int. J. Smart Nano Mater. 2019, 10, 205; b) J.-W. Su, W. Gao, K. Trinh, S. M. Kenderes, E. Tekin Pulatsu, C. Zhang, A. Whittington, M. Lin, J. Lin, Int. J. Smart Nano Mater. 2019, 10, 237.

[12] a) A. Mitchell, U. Lafont, M. Holýriska, C. Semprimoschnig, Addit. Manuf. 2018, 24, 606; b) M. Zarek, N. Mansour, S. Sharipa, D. Cohn, Macromol. Rapid Commun. 2017, 38, 1600628.

[13] a) I. Garces, C. Ayarici, Sens. Actuators, A 2020, 301, 111717; b) S. Akbari, A. H. Sakhaii, K. Kowsari, B. Yang, A. Serjejou, Z. Yuanfang, Q. Ge, Smart Mater. Struct. 2018, 27, 065027; c) H. Wu, O. Wang, Y. Tian, M. Wang, B. Su, C. Yan, K. Zhou, Y. Shi, ACS Appl. Mater. Interfaces 2020, f) C. Zhang, D. Cai, P. Liou, J.-W. Su, H. Deng, B. Vardhanabhuti, B. D. Ullery, S.-Y. Chen, J. Lin, Acta Biomater. 2021, 122, 101.

[14] a) J. Kim, J. Moon, Electrochem. Solid-State Lett. 2005, 8, 310.

[15] a) A. Bessonov, M. N. Kirikov, D. I. Petukhov, M. Allen, T. Ryhänen, M. J. Bailey, Nat. Mater. 2015, 14, 199.

[16] a) D. Li, W. Y. Lai, Y. Z. Zhang, W. Huang, Adv. Mater. 2018, 30, 1704738; b) H. Deng, C. Zhang, K. Sattari, Y. Ling, J.-W. Su, Z. Yan, J. Lin, Adv. Mater. Interfaces 2020, 13, 12719.

[17] M. Bohm, A. Ullmann, D. Zipperer, A. Knobloch, W. H. Glauert, W. Fix, presented at 2006 IEEE Int. Solid State Circuits Conf.-Digest of Technical Papers, San Francisco, CA, USA, February 2006.

[18] a) M. Aliyev, K. Stack, H. Lipson, Nat. Commun. 2017, 8, 596.

[19] X. Wan, L. Luo, Y. Liu, J. Leng, Adv. Sci. 2020, 7, 2001000.

[20] a) J. A. Lewis, J. E. Smay, J. Stuecker, J. Cesarano, J. Am. Ceram. Soc. 2006, 89, 3599; b) J. A. Lewis, Adv. Funct. Mater. 2006, 16, 2193.

[21] L. Li, Q. Lin, M. Tang, A. J. E. Duncan, C. Ke, Chem. – Eur. J. 2019, 25, 10768.

[22] J. A. Lewis, G. M. Gratson, Mater. Today 2004, 7, 32.

[23] S.-Z. Guo, F. Gosselin, N. Guerin, A.-M. Lanouette, M.-C. Heuzey, D. Therriault, Small 2013, 9, 4118.

[24] J. Lee, H.-C. Kim, J.-W. Choi, I. H. Lee, Int. J. Precision Eng. Manuf.-Green Technol. 2017, 4, 373.
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