

Digital manufacturing of functional materials for wearable electronics

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Abstract

Emerging development of functional materials provides promising solutions for wearable electronics, given their outstanding electronic, optical and mechanical properties. Digital manufacturing has the ability of rapidly building computer designed electronic devices from scratch, and provides tremendous opportunities for wearable electronic devices. This review summarizes the recent advances of digital manufactured wearable electronics devices, from the perspectives of functional materials, manufacturing methods and wearable electronic applications. The opportunities and challenges for digital manufactured wearable electronics are also discussed for this exciting field.

Introduction

Functional materials with outstanding properties are being rapidly developed for interdisciplinary applications. Organic semiconducting materials with higher mobility, lightweight, flexible and stretchable features are booming, providing better performances for semiconducting devices.¹ Metallic and alloy nanoparticles are also important materials with selective electrochemical sensitivities towards several key molecules in human respiration.² The combination of functional materials and other supporting materials can be used for novel

applications, such as the triboelectric nanogenerator (TENG).³ These large families of functional materials provide novel solutions for practical wearable electronic devices.⁴

The wearable electronics area is rapidly developing due to the high demand of personalized health care systems, which is the result of the emerging challenges from the aging society. Wearable sensors for physiological status monitoring, wearable power systems and human-computer interfaces are currently being emerging explored.⁵ Lightweight, flexible, stretchable, and comfortably wearable electronic devices are needed for this exciting field, as illustrated in Figure 1.

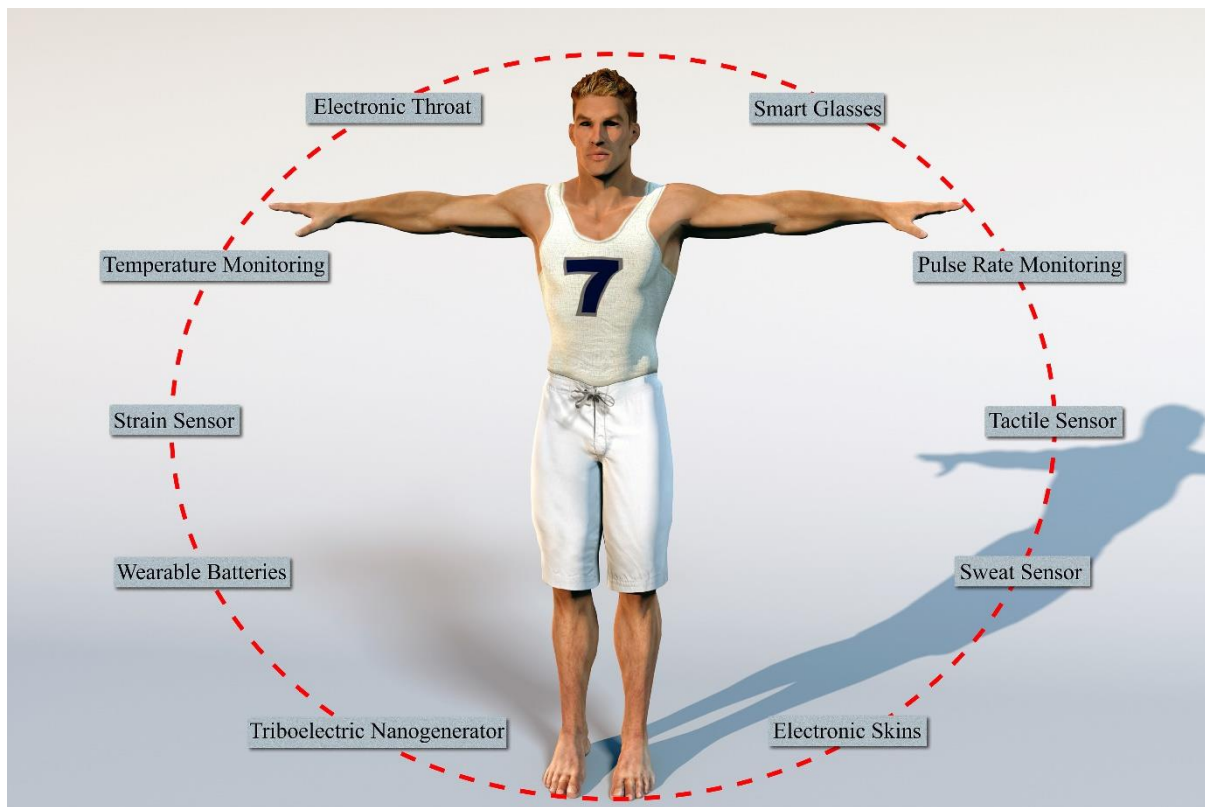


Figure 1 Illustration schemes of emerging wearable electronic devices.

Due to the large dissimilarities between different people, there are extremely high requirements for customized designs of devices for the wearers. Although traditional manufacturing methods, such as photolithography based semiconducting production lines, can produce high performance electronic devices, the lack of customized designs hinders the wearable

comfortability and limits wider applications. Personalized wearable devices with task-specific applications in distinct sizes are required for implementing wearable electronic applications.

Digital manufacturing methods can provide the customized designs for each of the fabricated modules. The computer blueprints can be directly fabricated into designed shapes and sizes through 3D printing, inkjet printing, laser writing and other digital manufacturing methods.⁶ The use of digital manufacturing methods of functional materials has been rapidly developed in the past five years, especially for wearable electronic applications. Although there have been several reviews on functional materials of wearable electronics, the digital manufacturing aspects of the emerging materials have not been thoroughly reviewed for wearable electronic applications. In this review, the key aspects of the digital manufacturing of the functional materials for wearable electronics are systematically discussed. The principles, restrictions and applications of various digital manufacturing methods are comprehensively discussed, considering the requirements of the functional materials for high performance wearable electronic devices. Meanwhile, the limitations and challenges are also discussed to spotlight the future developments in this new field.

2. Digital manufacturing of functional materials

Digital manufacturing provides an important means for transforming digital blueprints into real world samples. The process of digital manufacturing includes the design, fabrication and assembly processes. At the beginning, the blueprints of the materials are designed as patterns using 2D or 3D software.

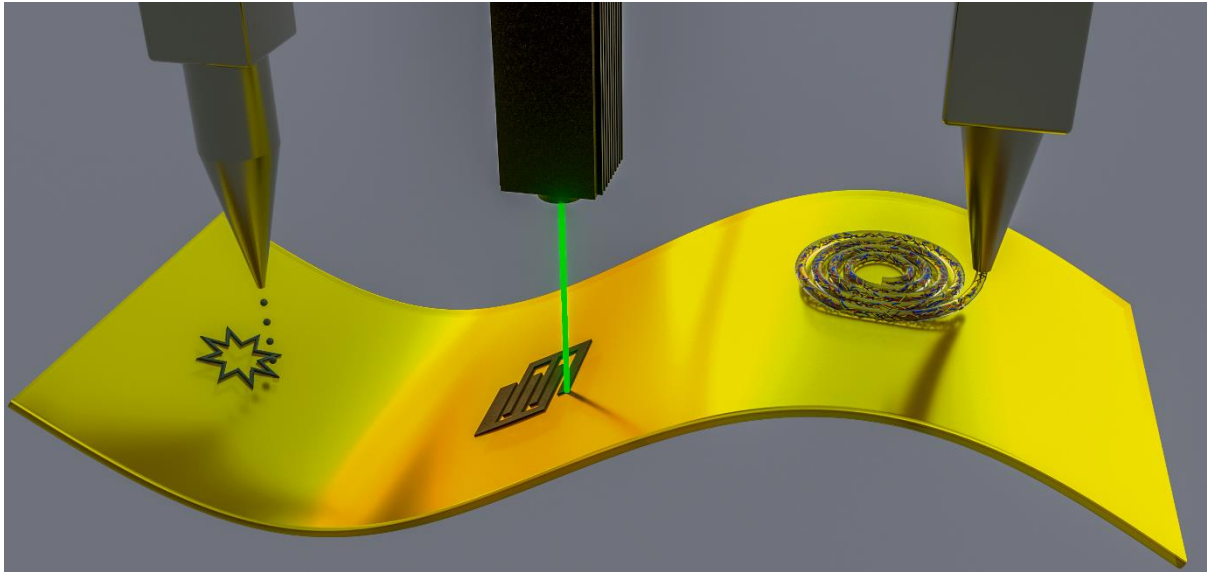


Figure 2 Illustration of digital manufacturing of flexible electronics, from left to right: inkjet printing, laser scribing and 3D printing.

The pixel sizes, widths and lengths of the product can be customized to meet the requirements of end-users, which is more advanced than the traditional fixed-mask based mass produced products of the same sizes. Digital manufacturing methods such as using inkjet printing, laser writing or 3D printing, are adopted to create wearable electronic components with functional materials, as shown in Figure 2. Finally, the fabricated devices can be assembled as integrated devices. The major methods of digital manufacturing are introduced below:

Inkjet printing

Inkjet printing is a digital manufacturing method for depositing functional materials onto the desired locations, as designed by the computer. It is a powerful method for patterning functional materials into wearable devices on almost all types of substrates. Other than using carbon ink for printing texts and figures, more types of functional materials can be printed for particular applications.

There are several components within inkjet printing, including the inkjet printing head (also called nozzle), cartridge, substrate and ink, as shown in Figure 3. The integrated systems can

transform the designs from the computer to different patterns of materials on various types of substrates.

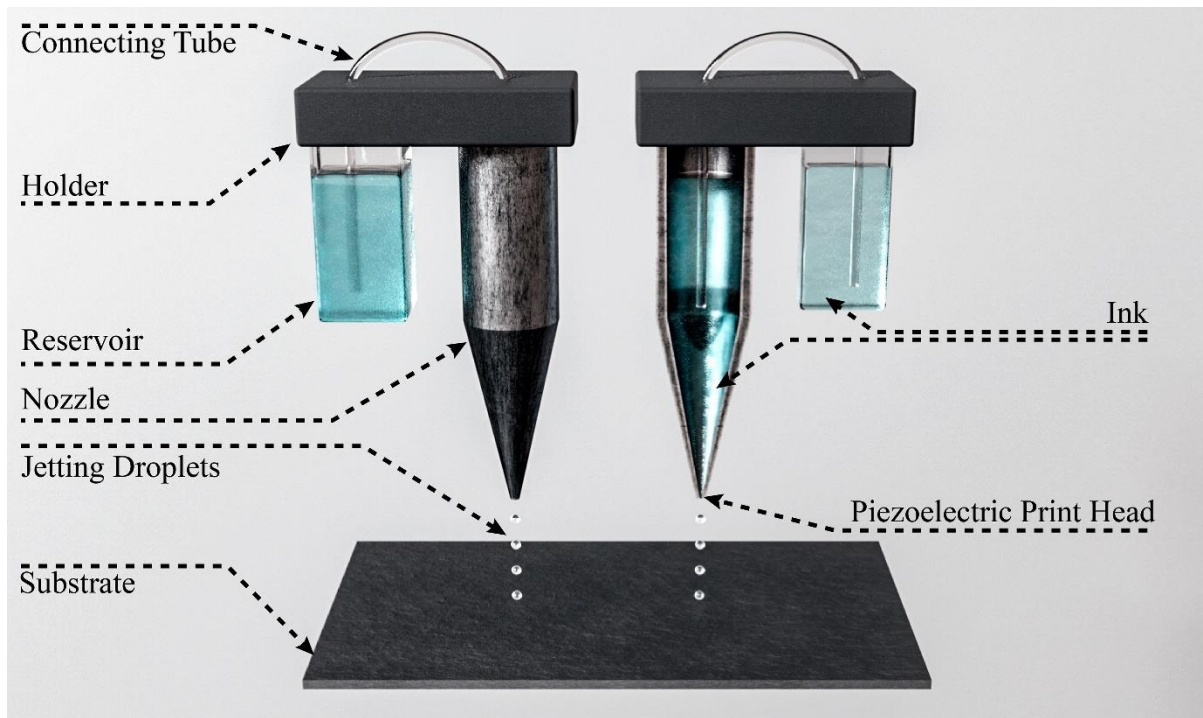


Figure 3 Components within the single-nozzle inkjet printing system for deposition of functional materials for electronic devices.

The cartridge is the container for the functional materials before the deposition through the nozzles. The inner surfaces of the cartridges are usually coated with polymer, so the types of solvents need to be considered with the potential reactions of the coating, so as to prevent contamination or failure of cartridge. For a strong polar solvent such as dimethylformamide (DMF), dimethyl sulfoxide (DMSO), it is recommended to use glass-based cartridges instead of polymer ones. The ink should be filtered with high density filters (usually with 0.4 or 0.225 micrometer pore size) before pouring into the cartridge, to prevent potential clogging issues. The potential chemical reaction of the solvent and the filter materials should be considered to prevent unwanted doping.

The inkjet printing nozzles are commonly classified into two categories: one is piezoelectric and the other one is thermal jetting nozzle. The thermal jetting head is the mostly and widely

used for office printers, due to its ease of fabrication and low cost. By locally heating the ink, the thermal expansion can create the momentum for jetting the ink onto the desired locations. However, as the temperature increases, it might be challenging for the deposition of some functional materials. For one aspect, the functional materials might break down or significantly change their properties, so it is not suitable for some temperature sensitive materials. In another aspect, the solvent might dry faster at the increased temperature, and the surface tension and viscosity vary significantly due to the higher temperature, so the jetting properties might change drastically. In contrast, the piezoelectric nozzle won't heat the ink significantly, so it would be more appropriate for the testing and depositing of custom inks for functional materials. The state-of-art printing heads are made of an array of nozzles instead of a single nozzle, as shown in Figure 4, so rapid deposition of functional materials onto different substrate areas simultaneously is possible with properly designed paths.

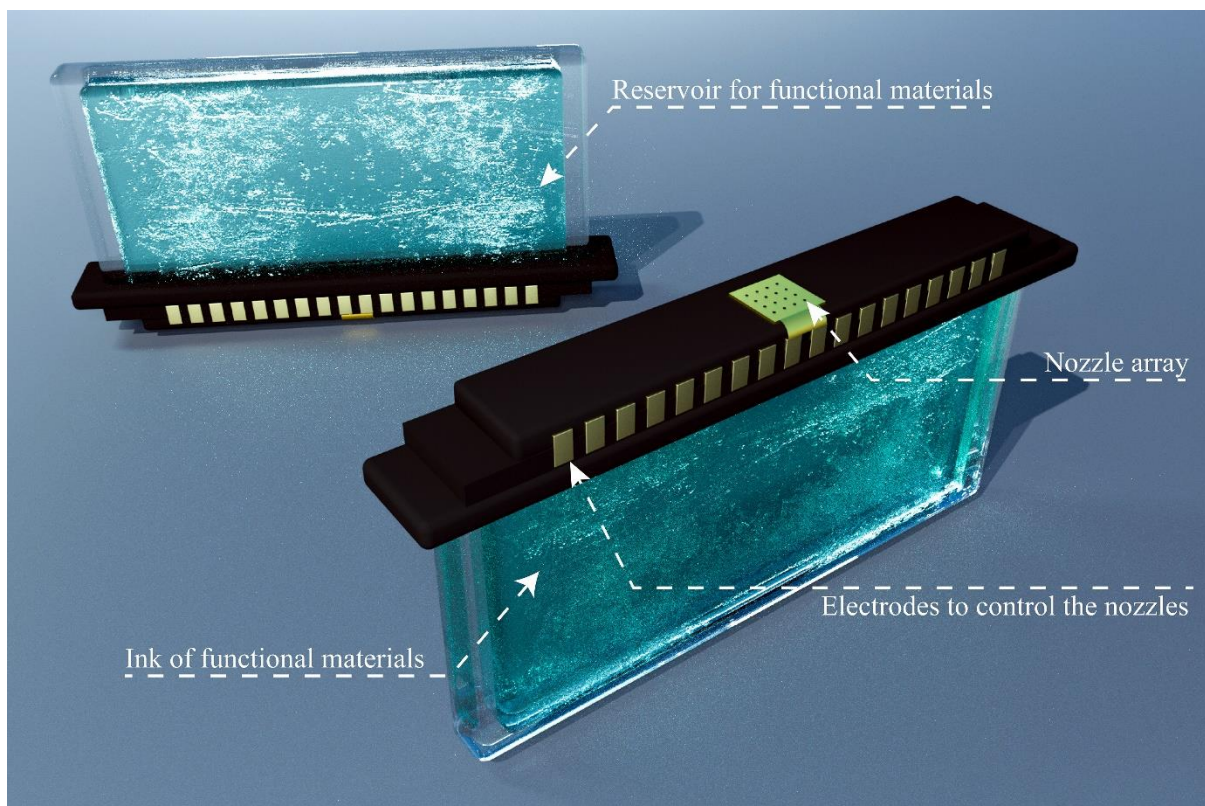


Figure 4 Inkjet printing with array of nozzles for simultaneously depositing of functional inks.

The relative movement of the nozzles and the substrates are controlled automatically using a microcontroller, which can be programmed through the computer designed paths. Both the movement of the nozzles and the platforms which hold the substrate can realize the deposition of functional inks onto different locations of the substrates.

The substrates for printing wearable electronics are usually flexible. Polymer substrates such as polyethylene terephthalate (PET), polyimide, polycarbonate (PC), polypropylene (PP), paper and polydimethylsiloxane (PDMS) are commonly used. The potential dissolving of the substrate materials with the functional ink needs to be considered. For high polar solvent, inert substrates should be used, such as polyimide or PDMS, considering the potential interaction between the solvent and the substrate.⁷

The wetting properties of the substrates should also be considered. Poor wetting properties can result in printing failures of the functional materials onto unwanted locations. For printing functional materials on flexible substrates, the wettability of the substrates and the solvent need to be considered together. For printing with aqueous ink, hydrophobic or superhydrophobic substrates should be avoided or modified. 3-Mercaptopropyltrimethoxysilane (MPTMS) can be added onto the PDMS surface to wet the nanoparticles based aqueous inks.⁸ UV ozone treatment is an efficient method for wetting the substrate by creating oxygen bindings on the surfaces of the substrates.⁹ For printing with oil-type organic solvents, oleophobic or superoleophobic substrates need pre-treatment before the printing. For example, some amphiphilic coatings can be deposited on the oleophobic surfaces to create an intermediate layer for ease of wetting the organic inks.¹⁰

For digital manufacturing of functional materials using inkjet printing, both the solution and the dispersion with satisfied distributions are feasible. Solution-based ink, considering the potential interaction of the inner surface of the cartridge and the substrates, can be used for

inkjet printing. For example, metallopolymer can be used for inkjet printing on glass and polyimide substrate with DMF as substrates. The dispersion inks with well controlled distributions can also work with inkjet printing systems. The first demonstrated inkjet printing of functional materials was performed with gold ink for fabricating a micro-electromechanical system (MEMS) fabrication using a self-developed inkjet printing system.¹¹ The inks can be either filtered or centrifugally separated to avoid the potential clogging of the nozzles during printing.

The temperature of the substrate also plays a key role for determining the final resolution of printed functional materials, and the solvent of the ink should be dried before actual usage. The drying process of the ink can prevent further movement of the ink on the substrate. Since most of the substrates for wearable electronics are flexible, the ink might lose control if it is still flowable on the substrates. By increasing the substrate temperature, the drying rates of the solvent can be increased, in order to decrease the free flow of ink on the flexible substrates. However, the breakdown temperature of the polymer substrate should be considered. For example, the PET substrate melts at 110 °C, so the substrate temperature for PET cannot be higher than this.

Laser manufacturing

Advanced laser manufacturing provides an alternative approach for fabricating wearable electronics with functional materials. Laser beams are usually scanned with either mechanical moving heads or by Galvo scanning systems, which can scan the laser beam at very fast speeds. State of the art lasers have wide power distributions, from nanowatts to megawatts, making the process highly scalable for mass production. Lasers also have widespread wavelength (as shown in Figure 5(a)), from deep (ultraviolet) UV to far infrared (IR), suitable for manufacturing materials with different wavelength dependent absorptions. The main

applications of laser manufacturing are in the three categories: laser cutting, laser reduction and laser printing.

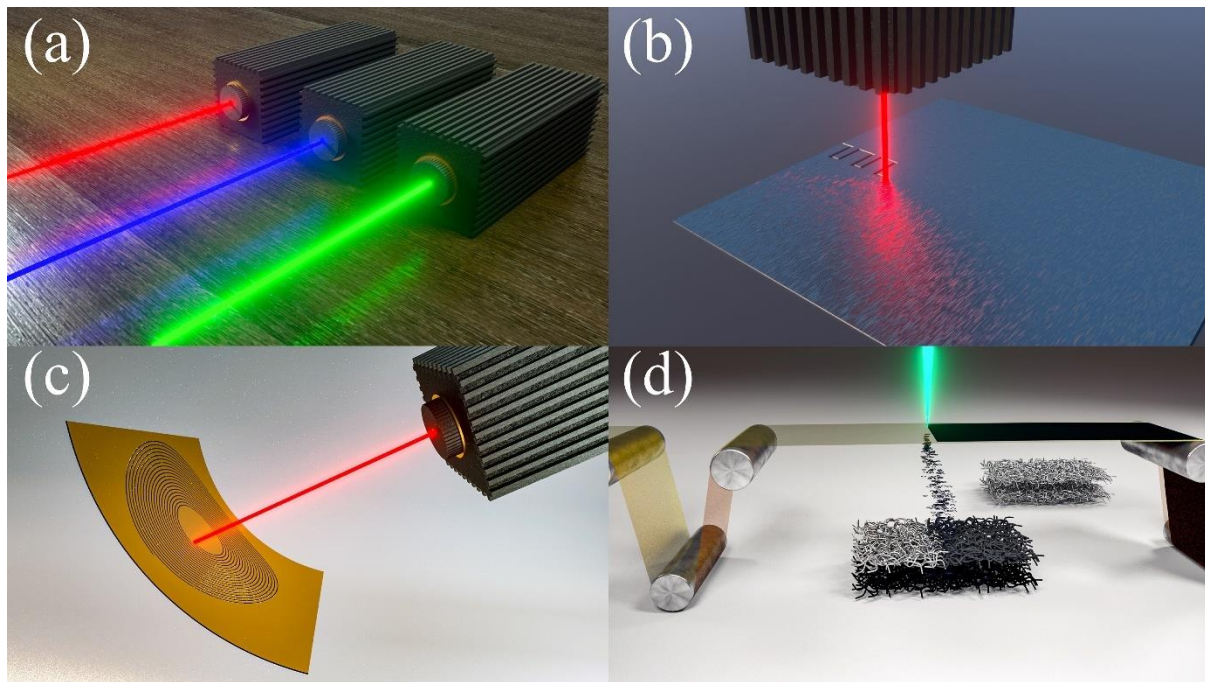


Figure 5 Laser manufacturing for wearable electronic applications. (a) Laser with different wavelengths. (b) Laser cutting. (c) Laser induced graphene on polyimide (d) Laser induced forward transfer of graphene.

Laser cutting utilizes the high-power density of a focused coherence laser beam, and physically vaporizes the unwanted materials with high temperature heating. The remaining patterns can therefore be used as functional components for wearable electronic applications, as shown in Figure 5(b). Any pre-coated materials can be removed by using the appropriate laser parameters, such as laser frequency and intensity. The main challenge with the laser cutting is its interaction of the substrate materials. The laser energy can be absorbed by the substrate, depending on its optical transmission rate. If the absorption of laser energy is significantly low for the substrate and high for the functional materials, the laser will selectively pattern the structures and not noticeably have influence on the substrate.

Laser reduction is an emerging technique for fabricating functional materials for wearable electronics. Laser induced graphene and laser reduced graphene are the main applications of

laser reduction, as shown in Figure 5(c).¹² Due to the relative high absorption of graphene oxide for infra-red energy, the oxygen-carbon bonding can be selectively reduced by the absorbed laser energy.¹³ As a result, the laser beam can photochemically reduce the graphene oxide and form the desired structures along the laser writing path. In 2014, it was also experimentally found that commercial polymer, such as polyimide, can be directly converted to laser-induced graphene by using laser writing.^{14, 15} This work has been successfully repeated in numerous research studies with applications for supercapacitors, biosensors, mechanical actuators, etc. The rapid heating of the aromatic carbon rings can form graphene structures, an effect that was also realized by electrical heating of different carbon sources. So the photothermal effect of the laser can be utilized to create graphene patterns on selective locations.

Laser printing is an alternative approach for creating functional materials with desired shapes, mainly based on the laser-induced forward transfer mechanism.¹⁶ By focusing on a donor layer with precursor materials, the functional materials can be selectively transferred onto the acceptor substrate. The transfer energy can either be the momentum from an ultra-short laser pulse or the sacrificial materials which can be photothermally ignited.^{17, 18} Femtosecond, picosecond and nanosecond lasers have all been demonstrated for the laser transfer of functional materials with various applications.¹⁹⁻²¹ The authors first demonstrated that the ABS can be used as a sacrificial material for the deposition of copper precursor materials using a continuous wave laser at 405 nm wavelength.²² Later, they successfully demonstrated that the 1064 nm continuous wave (CW) laser can simultaneously photochemical synthesis and print graphene onto other substrates, as shown in Figure 5(d).²³ In theory, almost any condensed matter materials, with proper melting temperature can be printed using laser induced forward transfer (LIFT).

3D printing

3D printing is an additive manufacturing process for the deposition of materials at designed locations. The design model could be built by the modelling software or 3D scanner, which will include the corresponding coordinates of the model. The three-dimensional coordinates will be converted into two-dimensional data for each layer by a process named slicing, and a machine paths for the printer nozzle is designed accordingly. Then data is transferred to the microcontrollers of the 3D printer, and the melted functional materials are extruded through the nozzle to fabricate the designed model. The mighty ability of 3D printing to selectively place the functional materials onto the desired locations, enables the constructions of the real-world geometries, as shown in Figure 6. Commercial 3D printers are widely available across educational institutes. The challenges of 3D printing for electronic devices are limited by the choice of printable materials.

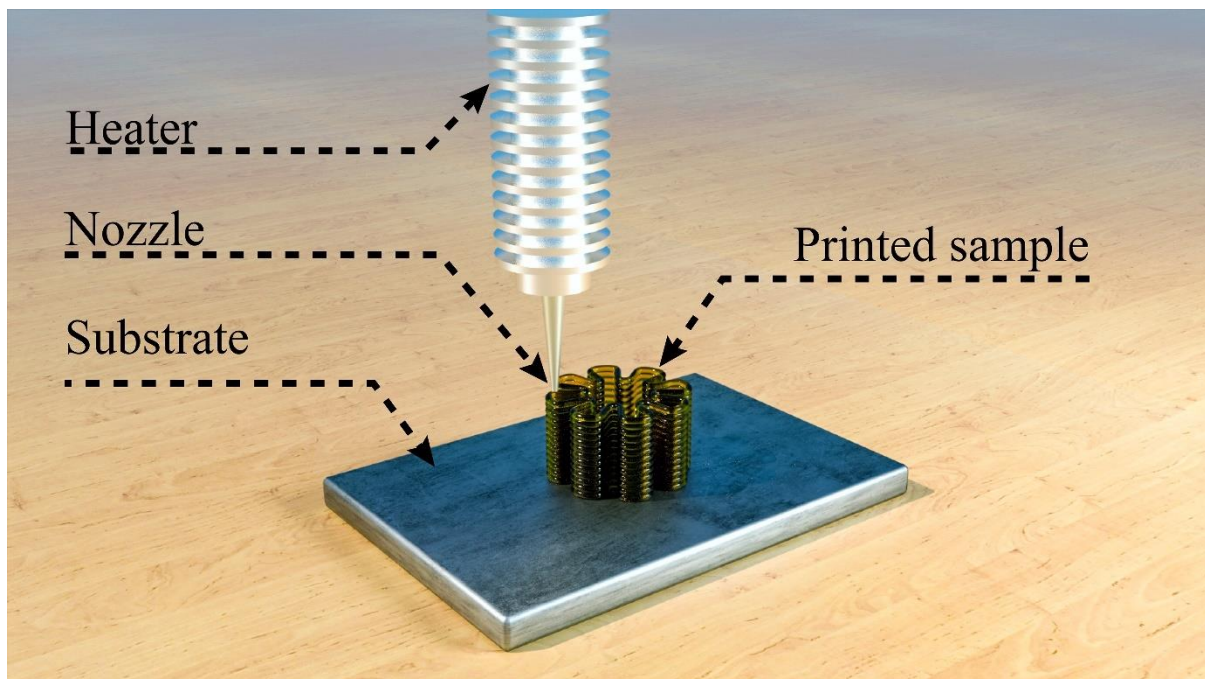


Figure 6 Components for 3D printing nozzle.

3. Functional materials for wearable electronics

There are two aspects of the material requirements for wearable electronic applications fabricated by digital manufacturing methods. One is to be compatible for wearable electronics applications, and the other is to match with existing digital manufacturing methods.

For wearable electronic applications, there are a few constraints. First, the material should be non-toxic to humans through airborne or physical contact. So highly toxic materials must be avoided for direct use in wearable electronic devices, even though they might possess high performance in applications. For example, nickel oxide nanoparticles are high-performance semiconducting materials. However, due to its high toxicity, they should not be used directly in wearable electronic devices.²⁴

Secondly, the comfortability of the functional materials should also be considered. For the long-term wearable devices, it is crucial that the users feel comfortable during usage. Anti-inflammation should be considered for the materials that can directly contact with the skin. So the materials of the substrates, which usually directly contact the skin, should have good comfortability and be chemically safe. One of the most important factors is the stretchable possibility of the material. It is significant to have the extensibility of the finalized product remained in the range of elastic tensile strain for the skin, 15% epidermal strain as reported.²⁵ Two prevalent methods were commonly reported, one was to provide elongation property by the chemical reactions and the structure construction of various materials,²⁶ and the other one was to assemble the intrinsic stretchable material with other elastomeric materials.²⁷⁻²⁹ In addition, kirigami structures were frequently adopted for wearable application.^{30,31} At the same time, the user habits toward long-term usage should also be considered. Paper based wearable electronics are booming due to its low cost and ease of manufacturing.³² However, current devices made of paper are not durable enough for long-term usages.

Thirdly, the mechanical and electronic properties of the materials should not degrade drastically when they are applied used as wearable devices. The flexibility, stretchability and light-weight features should be considered with the performance. All after, the usability of the devices is essential, for example in replacing a large-scale equipment in hospital.

For digital manufacturing, the materials should also be compatible with at least one of the existing methods, such as inkjet printing, laser manufacturing, and 3D printing.

3.1 Functional materials for inkjet printing

For inkjet printing or other wet printing methods, the surface tension and viscosity of the ink are key parameters to determine whether the selected type of materials can be printed. The viscosity of the ink is determined simultaneously by the solvent and the solute for the solution, and by the solvent and the dispersed materials for the dispersion. By adding in sodium dodecylbenzenesulfonate (SDBS), the viscosity of the single-wall carbon nanotube (SWCNT) in water can be maintained at 20 cp at 1 s^{-1} for printing electrodes for supercapacitor applications.³² By adding 5% dimethyl sulfoxide (DMSO) in water, the viscosity of the poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) ink can be maintained at 100 cp at 1 s^{-1} , which can be used for printing circuits on wearable textiles.³³

Some of the solution-based inks can be printed directly through the nozzles, such as Zinc acetate dihydrate in ethanol for printing the precursor for the ZnO thin film.³⁴ However, most of the emerging novel materials cannot be dissolved in solvents, so their sizes and distributions within the inks need to be finely controlled. Some extra procedures such as ball milling, sonication and surfactant additions are common for pre-treating the inks before printing, for producing well-dispersed inks, as shown in Figure 7. Graphene at 3.5 mg/mL density can be printed in a composite solution of 85% cyclohexanone and 15% terpineol with added ethyl cellulose 5% in toluene/ethanol 80:20, for printing H_2O_2 sensors.³⁵ Few-layer graphene with

density of 0.8 g/cm^3 and viscosity of 2.3 mPa s mixed with PVP and IPA was inkjet-printed for thermoelectric applications.³⁶ For supercapacitor applications, rGO/MoO₃ composites³⁷, carbon quantum dots/graphene oxide inks³⁸ can also be dispersed in the water for inkjet printing.

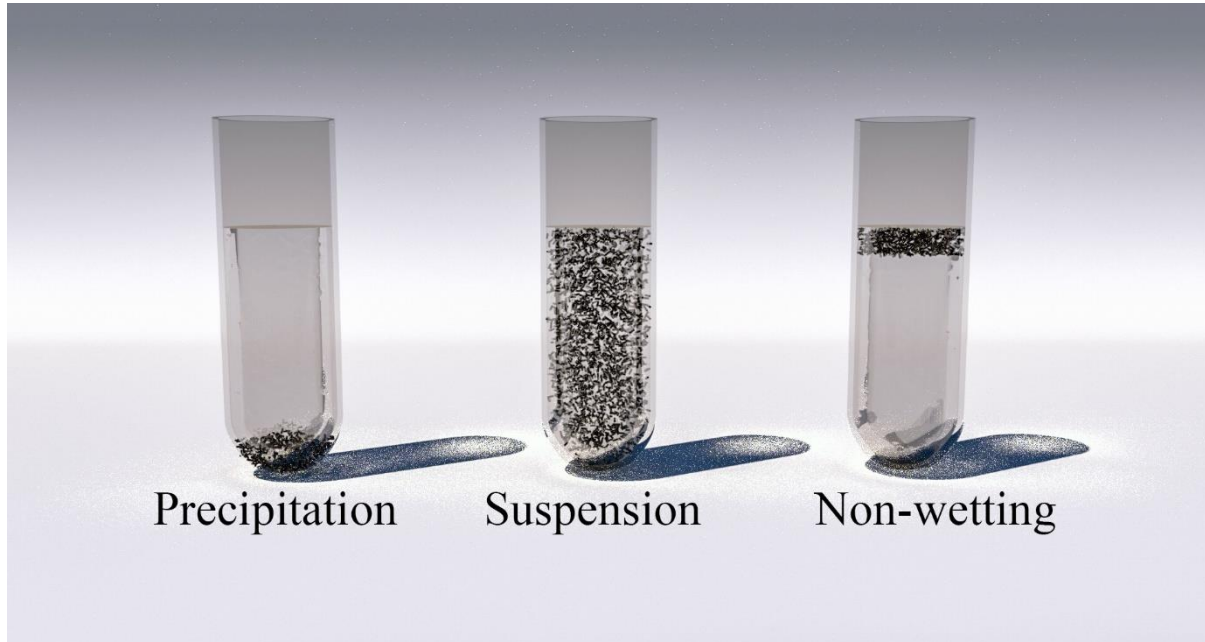


Figure 7 Different states of distribution of functional materials within the solvent, from left to right: precipitation (poor), uniform suspension (good), and non-wetting (poor).

With properly designed physical properties of the inks, integrated systems can also be inkjet printed.³⁹ By sequentially printing polyvinyl chloride (PVC) and 2,7-dioctyl[1]benzothieno[3,2-b][1]benzothiophene (C8-BTBT/PS) in anisole, the organic transistor at a high gain and low voltage can be fully inkjet printed.⁴⁰ By sequentially inkjet printing MnO₂/rGO/PEDOT:PSS as supercapacitor electrodes and SnO₂ as the gas sensor, a self-powered gas detection systems can be built.⁴¹ Various components within the organic transistors can also be fully inkjet printed, with 10 mg/mL poly(vinylidene fluoride)-co-hexafluoropropylene (PVDF-HFP) in N-methyl-2-pyrrolidone as gate dielectric, 0.03 mg/mL SC-SWCNT dispersed in toluene at channel layer, PEDOT:PSS in water with added DMSO and SDBS as source and drain, and silver for conducting pads.³⁸ Compared to 3D printing, inkjet printing method had significantly higher resolution, which can be selected for

manufacturing compact devices. Due to the less use of additive composite material for extruding through 3D printing nozzles, the inkjet printed structures usually possessed higher conductivity.^{42, 43}

3.2 Functional materials for laser manufacturing

Compared to inkjet printing, laser manufacturing has a higher tolerance in the choice of materials, because it is free from the clogging issues of the inkjet printing nozzles. Carbon rich precursors, such as polyimide, graphene oxide, etc., as shown in Figure 8, have been studied as for generating laser induced graphene.⁴⁴ Metal-containing organic materials can also work as the precursors for producing metallic nanoparticles.⁴⁵ Laser induced forward transfer is capable of printing many different kinds of materials, even with live cells, due to its low temperature process.⁴⁶ However, the phase change temperatures are crucial for determining the laser printing quality. If the melting temperature is too low, it is challenging to print using a laser. For example, it is impossible to print ice with a laser. Although printing with liquid inks have been demonstrated using LIFT, most materials are laser printed in a solid or half-melted phase.⁴⁷

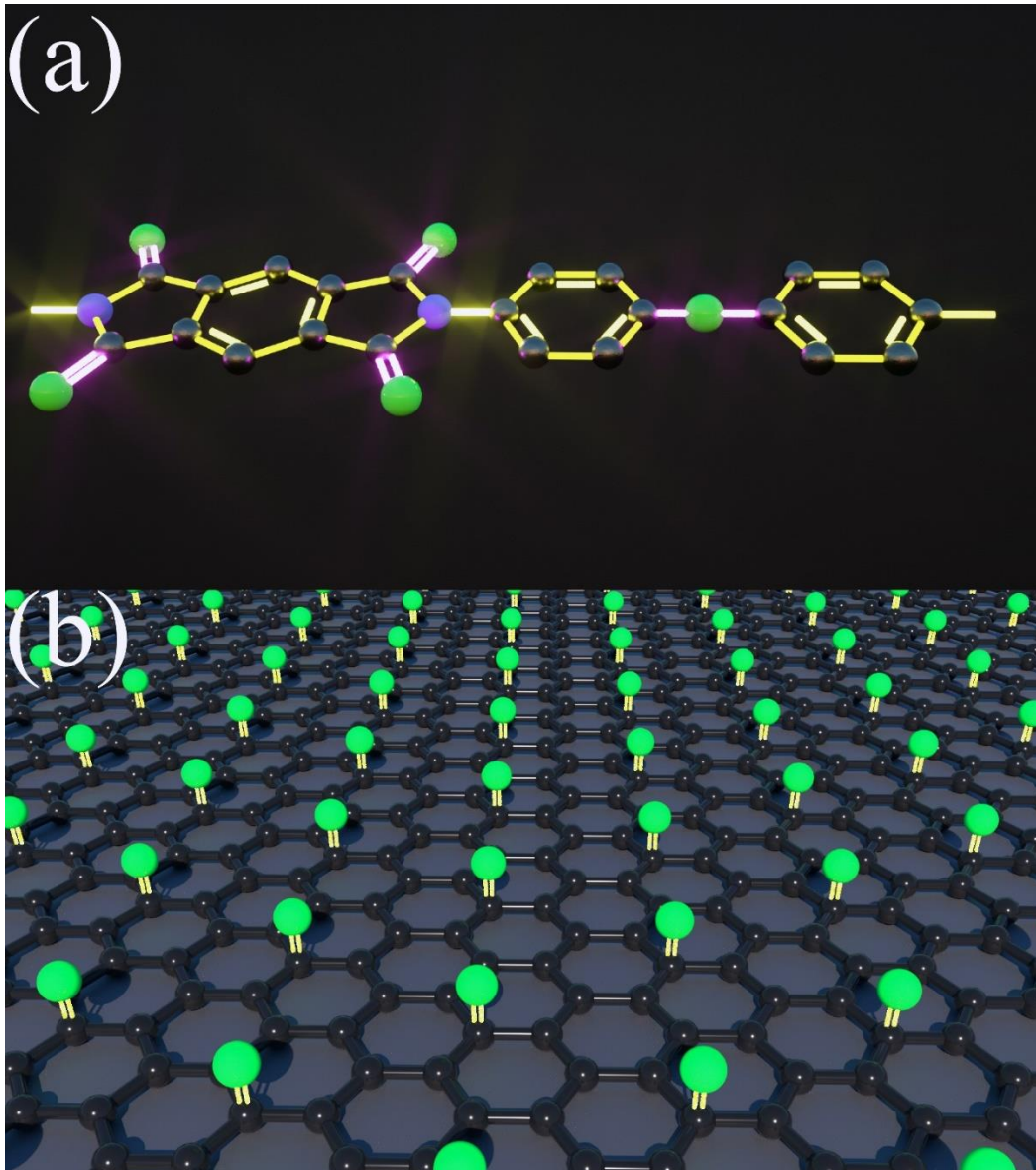


Figure 8 Precursor materials for laser synthesis of graphene. (a) Polyimide molecule (b) Graphene oxide.

3.3 Functional materials for 3D printing

It is challenging to 3D print existing materials directly, except for some high viscosity polymers, such as silicone elastomers.⁴⁸ Usually, a polymer will be mixed with the functional materials to form the composite precursor materials. Ecoflex elastomers are frequently reported to aid the 3D printing of other functional materials, as illustrated in Figure 9. By mixing the Ecoflex sets with MWCNT, a commercial 3D printer with a 0.4 mm diameter nozzle can be used for printing highly sensitive tactile and electrochemical sensors.⁴⁹ When the Ecoflex was mixed

with carbon nanoparticles, the composites could be printed with a 0.8 mm nozzle as flexible and stretchable electronic skins.⁵⁰ When the Ag flakes were mixed with Ecoflex, PDMS and MIBK solvent, the composites could be 3D printed as self-wiring ink with a 0.51 mm nozzle at a 1 mm/s speed.⁵¹ At the same time, PDMS was frequently wet and mixed with MWCNT as 3D printing ink, for stretchable electrodes⁵² and wearable microheater applications.⁵³ Silver particles were frequently 3D printed for wearable electronic applications, mixed with silicone,⁵⁴ poly(ethylene oxide) (PEO),⁵⁵ polylactic acid (PLA),⁵⁶ etc., as composite precursor materials. Although similar functional materials, such as MWCNT, can be fabricated using inkjet printing, the viscosity of the composite inks for 3D printing is significantly higher than that for inkjet printing. In addition, the inkjet printing had limited capability for fabricating high aspect ratio microstructures compared to 3D printing. For building wearable electronic components beyond two dimensions, 3D printing is recommended.

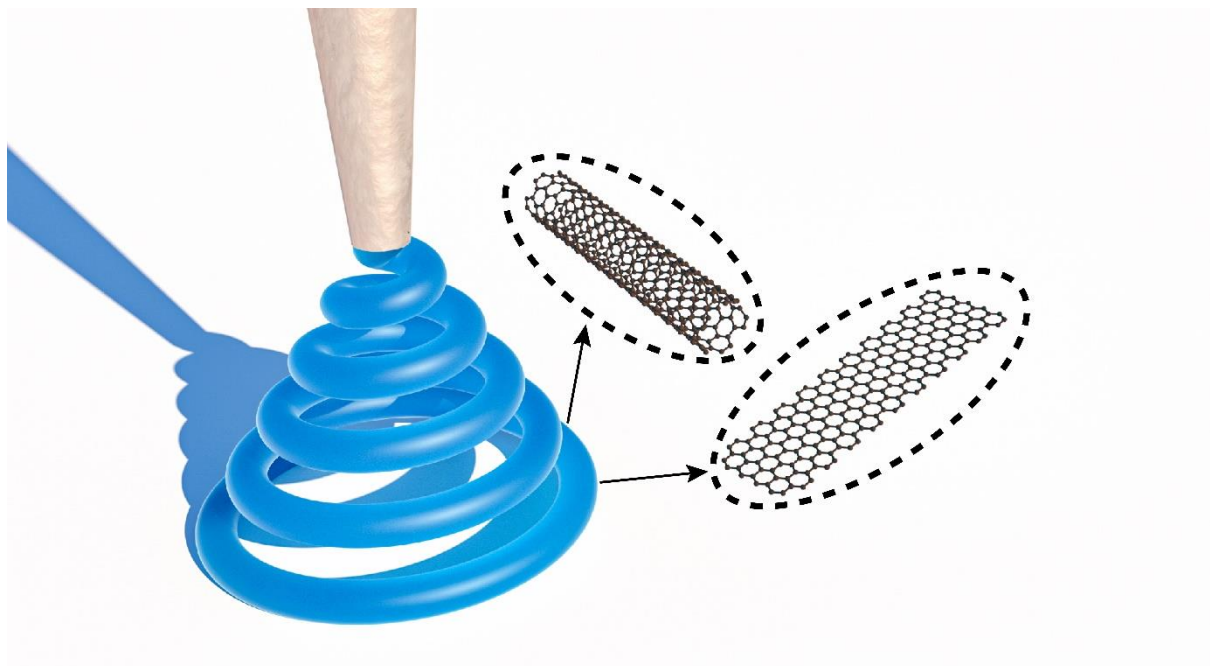


Figure 9 Mixing of graphene and carbon nanotube in the silicone for 3D printing.

Dry mixing is also capable of producing filaments containing functional materials as precursor for commercially available 3D printers. Through twin-screw extrusion, the MWCNT can be dry mixed with TPU, which can be printed by a commercial 3D printer at 220 °C at 20 mm/s

speed with a 0.8 mm nozzle, for highly elastic strain sensing applications.⁵⁷ The TPU can also be planetarily mixed with silver as the precursor for fabricating soft electronic devices.⁵⁸ The carbon black and homo polypropylene can be mixed using a single screw extruder, which can be printed at 230 °C using a 0.4 mm nozzle as a 3D printable electrically conductive filament.⁵⁹ The clobetasol propionate can also be mixed with PLA using a single-screw extruder, which can be used as filaments for 3D printing at 195 °C at 50 mm/s speed using a 0.35 mm nozzle, as a wearable personalized oral delivery device.⁶⁰

The functional materials can also be all-3D-printed as integrated systems. For example, silver nanoparticles, P3HT:PCBM and PEDOT:PSS with modified surface tensions and viscosities can all be printed using 3D printing with 0.1 mm nozzle for an integrated photodetector application.⁶¹

4. Wearable electronic devices from digital manufactured materials

4.1 Energy applications:

Electrical energy provides the source for active devices for wearable electronic applications. Energy storage and conversion devices can both provide energy for these devices fabricated by digital manufacturing methods.

4.1.1 Triboelectric nanogenerator

The energy generating/conversion devices can provide instant power, including triboelectrically generators, solar cells and fuel cells. The wearable triboelectric nanogenerator can convert the kinetic energy to electricity in a lightweight and flexible fashion. Several digital manufacturing methods have been demonstrated for fabricating TENG for wearable electronics, as shown in Figure 10. The inkjet printed TENG showed an output voltage as high as 124 V at a power density up to 608.5 mW/m², which could power a wearable electrowetting display and 51 LEDs.⁴⁸ 3D printed snow-based TENG demonstrated an open circuit voltage up to 8 V with

instant output power density up to 0.2 mW/m^2 and current density of $40 \text{ }\mu\text{A/m}^2$, which can work in snowy weather conditions.⁶² The laser induced graphene can also work as electrodes for the TENG with high open-circuit voltage up to 3,500 V, with a peak power over 8 mW as flexible wearable electronic energy sources.⁶³ These digital manufactured TENG device can work as instant power supplies with high peak powers and voltage for various electronic devices. To provide a constant electrical power, the TENG can be integrated with a power storage unit, such as a flexible micro-supercapacitor.⁶⁴ The strong customization ability of the digital manufactured TENG can support a wide range of applications for wearable electronics.⁶⁵ The key challenge of TENGs is their resistance towards stretching and twisting. Kirigami and other structures can be artificially designed, benefiting from the flexibility of digital manufacturing.⁶⁶

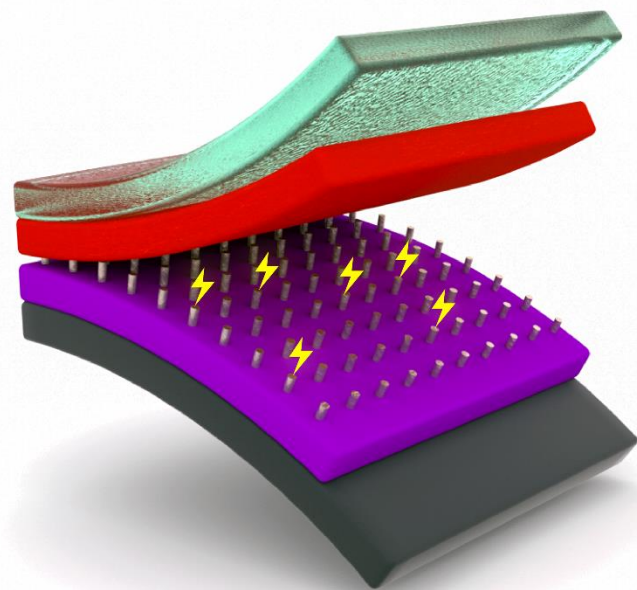


Figure 10 Illustration of nanostructured multilayer for TENG device.

4.1.2 Solar cells

Solar cells are efficient power conversion devices that can be used for directly providing electricity from sunshine illumination. Digital manufacturing provides great support for emerging solar cells, which benefit significantly from the solution processing methods. Highly efficient P₃HT/fullerene solar cells fabricated by inkjet printing were first demonstrated in 2008, with efficiency up to 3.5%.⁶⁷ Since then the indium tin oxide (ITO) free solar cells were frequently reported with inkjet-printed current collectors.⁶⁸⁻⁷⁰ Organic solar cells, inorganic solar cells and perovskite halide solar cells have all been successfully inkjet printed with high efficiencies.⁷¹⁻⁷³ Thick perovskite solar cells at 1.5 μm thickness with large grain sizes had a high power conversion efficiency (PCE) of 21% were successfully inkjet printed in a digital manufacturing manner.⁷⁴ At the same time, laser writing techniques also demonstrated useful applications for solar cell manufacturing, such as deposition, patterning, sintering and sealing.⁷⁵ In addition, the laser treatment is very efficient for promoting the crystallinity and efficiency for halide perovskite solar cells.⁷⁶ Large grains of perovskite with high efficiency up to 21.2% had been successfully demonstrated using ultra-fast laser writing.⁷⁷ These digital manufactured solar cells can provide an instant power supply under sunlight. When the energy storage devices are integrated with these solar cells, a sustainable power system is formed for long-term usage, which can provide a green energy source for wearable electronics.⁷⁸ The challenges of scalability can be properly solved by digital manufacturing methods, which provide promising opportunities for the emerging development of solar cell materials.

4.1.3 Fuel cell

The fuel cell is another method for generating electricity from multiple chemical resources. Inkjet printing of electrode materials has been widely studied for fuel cell applications. Microbial fuel cells with inkjet-printed polyaniline (PANI) gel as the anode can provide a 6.1 fold increase in current density, compared to unmodified carbon paper.⁷⁹ The cerium oxide decorated polymer nanofibers fabricated by inkjet printing can provide ultralong durability

with a voltage decay rate as low as 0.39 mV/h. Digital laser manufacturing is also an useful method for creating fuel cell electrodes. Additive laser manufactured electrodes with high current density up to 1.515 A/cm² and power density at 363 mW/cm² were demonstrated as stainless steel micro fuel cells.⁸⁰ The laser writing method can also create active porous carbon surfaces with enhanced catalytic performance of the embedded platinum nanoparticles for proton exchange membrane fuel cells.⁸¹ The printed microbial fuel cells with power density of 6.4 μW/cm² and current density of 52 μA/cm² proved excellent folding ability toward stretching and twisting.⁸²

4.1.4 Batteries

Compared to energy conversion devices, digital manufactured energy storage devices are usually effective for providing energy, without the need of considering the critical conditions for power generation. The wearable energy storage devices are usually classified into two categories: i.e. batteries with high energy densities and supercapacitors with high power densities.

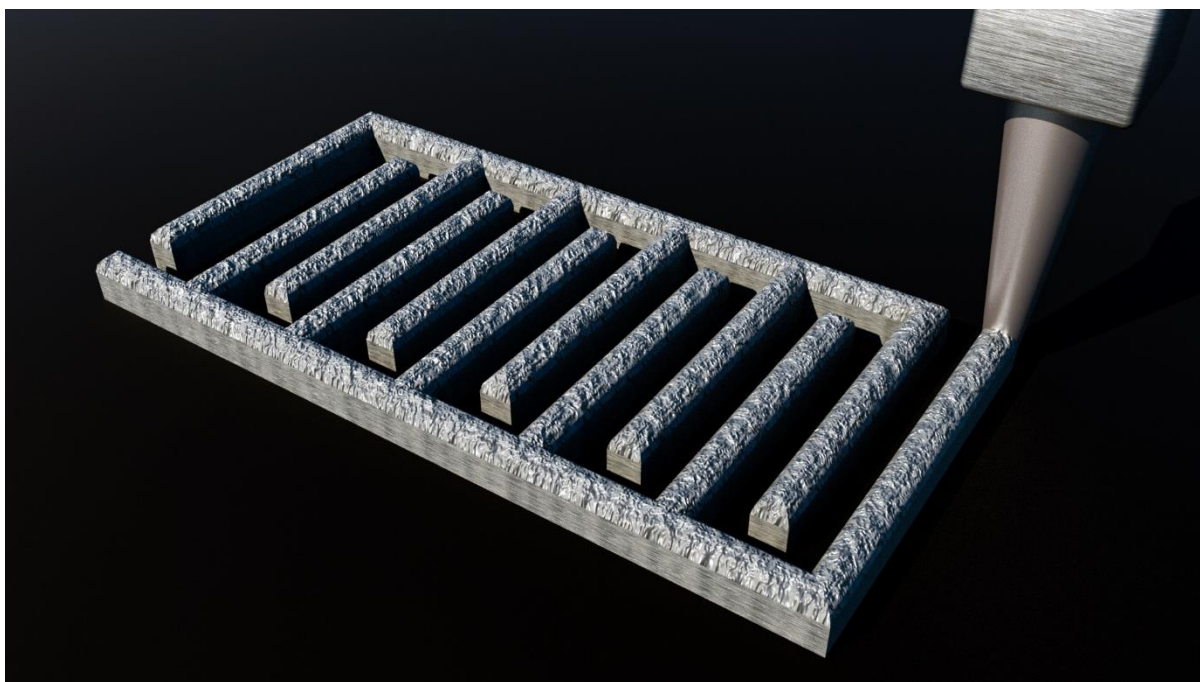


Figure 11 3D printing of electrode for energy storage.

Wearable batteries are emerging energy devices due to their high electricity capacities. The digital manufacturing methods are efficient for creating customized designed electrodes for batteries, as shown in Figure 11. The fiber-based lithium battery with specific capacity of 110 mAh/g at current density of 50 mA/g was 3D printed using a gel polymer.⁸³ The fused filament fabrication of lithium ion batteries with controlled LiClO₄ doping ratios were demonstrated for 3D printed wearable batteries.⁸⁴ Due to the excellent interpenetrating transmission paths and channels for electrons and ions from the 3D skeleton, high-loading lithium-sulfur batteries were also 3D printed, with 505.4 mAh/g after 500 cycles.⁸⁵ Laser machined PANI/carbon fiber with laser micromachined zinc anodes can be integrated into 3D printed rings for rechargeable batteries.⁸⁶ Laser lift-off can fabricate thin film flexible batteries used in smart lens with electrochemically active LiFePO₄ with 70 $\mu\text{Wh}/\text{cm}^2$ energy density for powering a glucose sensor for 11.7 h.⁸⁷ Laser induced graphene can also work as the anode for a lithium ion battery with 280 $\mu\text{Ah}/\text{cm}^2$ initial real capacity.⁸⁸ It had also been reported to possess a peak power density of 98.9 mW/cm^2 and energy density of 842 Wh/kg as electrodes for the Zn-air

batteries.⁸⁹ These wearable batteries are promising power resources with customized designed features.

4.1.5 Supercapacitors

The supercapacitor is an energy storage device based on the principles of electrical double layer capacitance and pseudocapacitance. Due to the non-intercalation nature of the electrochemical storage, the supercapacitors are usually capable of fast charging and discharging, which are very useful for short-duration and large power density applications. Due to the ease of fabricating artificial shapes, the digital manufacturing methods have been widely studied for patterning interdigital patterns for supercapacitors. Direct laser writing of polyimide can synthesize laser induced graphene, which is a low-cost and fast method for creating the interdigital electrodes of supercapacitors.¹⁴ The sulfonated poly(ether ether ketone) (SPEEK) can work as an alternative precursor material for creating flexible supercapacitors by direct laser-induced graphenization.⁹⁰ In-plane micro-supercapacitors have been fabricated using different laser sources, ranging from the CO₂ laser to the femtosecond laser.^{91, 92} Reduced graphene oxide and gold nanoparticles can also be laser written into micro-supercapacitors for wearable electricity sources.⁹³ Ultrathin laser processed micro-supercapacitors with 5.7 mWh/cm³ volumetric energy density was achieved for powering wearable LEDs.⁹⁴ The graphene oxide and black phosphorus quantum dots can also work as the precursor materials for directly writing the flexible planar supercapacitor using laser.⁹⁵ Different patterns and scales can be realized, demonstrating the scalability of laser writing for creating supercapacitors electrodes.⁹⁶ The highly porous inner structures and conductivities are the advantages of the laser manufactured functional materials.

At the same time, inkjet printing of different functional materials has also been widely studied for wearable supercapacitors. With CNT inter-welded Ag nanowires as electrodes, flexible

supercapacitors can be fully-inkjet-printed as flexible power sources.³² With solely printed 2D materials, the integration of circuits and capacitors have been demonstrated using inkjet printing for energy storage, resistor-capacitor low pass filters and transistors.^{97, 98} Beyond electrode materials, latex PVDF with PVA can be inkjet-printed as the dielectric media, realizing a high discharge energy density of 12 J/cm^3 at 550 MV/m with outstanding mechanical robustness and stability over time.⁹⁹ Inkjet printed RGO and MoO_3 nanosheets can work as all-solid-state electrodes for flexible supercapacitor applications, with maximum energy density of 2 mWh/cm^3 and power density of 0.018 W/cm^3 .³⁷ Among the reported inkjet printed ink, MnO_2 nanoink possess the highest specific areal capacitance.^{100, 101} The combination of laser writing and inkjet printing can also create flexible supercapacitors. By introducing pseudocapacitive cobalt molecules via inkjet printing, the areal specific capacitance of the interdigital laser scribed electrode can be improved by 45 times.¹⁰² As a result, the introduction of pseudocapacitive functional materials by inkjet printing can further enhance the capacitance of supercapacitors.

Meanwhile, 3D printing might not provide additional inner surface areas as electrical double-layer capacitance. However, it can create artificial shaped supercapacitor devices and increase the specific areal capacitances which can enhance the areal power density for wearable applications.¹⁰³⁻¹⁰⁵

4.2 Sensors:

4.2.1 Tactile sensors

Tactile sensors are sensing elements that can simulate the physical sensing of human skin. There are several types of tactile sensors, such as pressure sensors, strain sensors, and vibration sensors. The digital manufacturing of the tactile sensor for wearable electronics have been

evolving, as shown in Figure 12, for Virtual Reality (VR), Augmented Reality (AR), soft robotics and prostheses applications.¹⁰⁶ 3D printed tactile sensors have been developed for working on freeform surface at ambient conditions, for advanced bionic skin applications.⁵⁴ Capacitive sensors were 3D printed using multiwall CNT for highly sensitive tactile sensing.⁴⁹ Spider-web shaped tactile sensors can also be 3D printed with the aid of collaborate robotic arms using porous graphene.¹⁰⁷

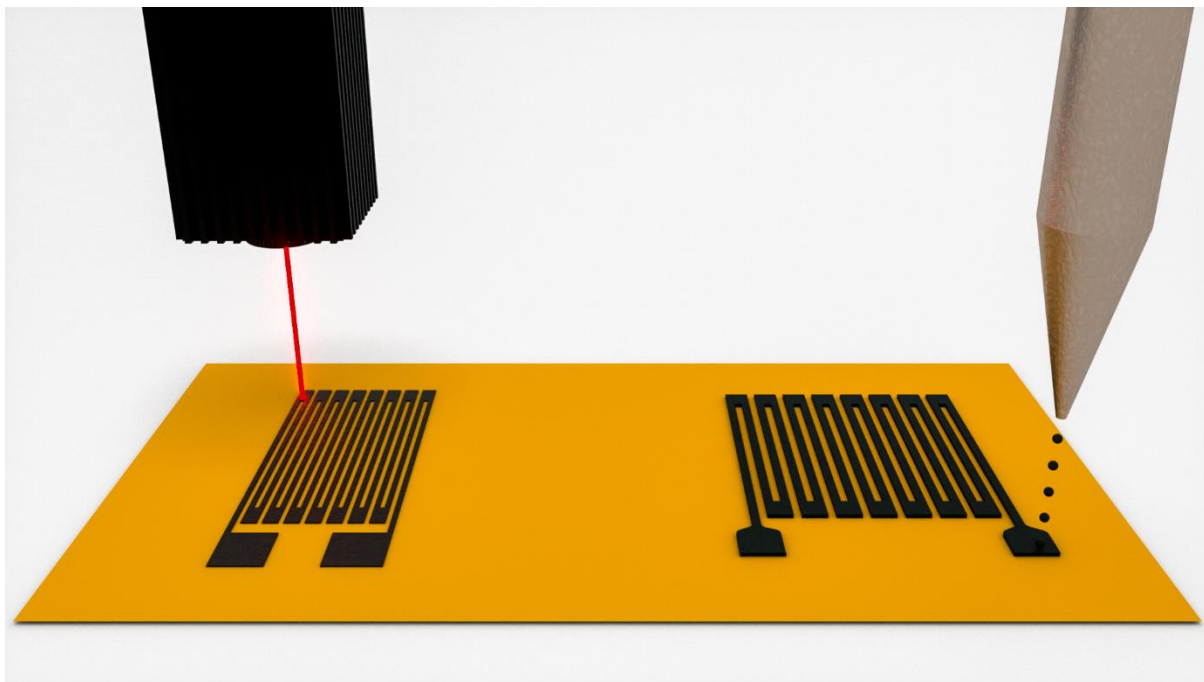


Figure 12 Digital manufacturing of strain sensor on flexible substrate. From left to right: laser direct scribing, and inkjet printing.

For strain sensor applications, many wearable applications have been fabricated using digital manufacturing methods. For 3D printing, the elastic strain sensors were fabricated using CNT and thermoplastic polyurethane composites with gauge factors as high as 176.⁵⁷ CNT and graphene composites were used as the vertically stackable strain sensors after 3D printing, with the gauge factor over 70.¹⁰⁸ With the addition of PDMS for the MWCNT, the 3D printed strain sensor carried excellent stretchable and flexible ability, with long term stability over 300 cycles.¹⁰⁹ When the silver nanoparticles were added to the 3D printed CNT, the fabricated

strain sensors had gauge factors up to 43,260 at 250% strain, fast response time at 57 ms and outstanding stability after 1000 cycles of stretching.¹¹⁰

Inkjet printing has also been explored for wearable tactile sensing applications. Graphene flakes and ZnO composites printed with commercialized inkjet printers had stretchability up to 30%.¹¹¹ The strain and pressure sensors were inkjet printed together as bimodal sensors, with gauge factor of 4000 and were stable over 4500 cycles.¹¹² The inkjet printed silver nanoparticles on PDMS can also worked as a resistive type pressure sensor, with sensitivity up to 0.48 kPa⁻¹.¹¹³ Aerosol jet printing was examined for building bandage-based strain sensors with good sensitivity over 700 repeated cycles, thanks to the post laser sintering of the printed nanoparticles.¹¹⁴ Laser writing has also been utilized for fabricating various strain sensors. The sputtered metal on polymer membranes can be selectively written by a laser as a strain sensor, with linear output up to 85%.¹¹⁵ After laser engraving on the CNT paper, the fabricated strain sensor showed a gauge factor over 42,000 at 150% strain and was highly linearly at large strains.¹¹⁶ With the laser induced graphene strain sensor, a gauge factor of 0.47 could be achieved by direct laser writing on polyimide.¹¹⁷

From the above comparison, it was found that the 3D printed strain sensors usually show higher stretchability compared to other methods, due to the consideration of using a fiber type composite. However, it is challenging to directly inkjet print fibers through the nozzle due to the clogging issues. So for inkjet printing strain sensors, it is important to consider introducing alternative fibers to enhance the stretchability.

4.2.2 Temperature sensors

High accuracy wearable temperature sensors are in high demand with the recent COVID-19 outbreak. The digital manufacturing of temperature sensors has been frequently reported in recent few years. Through 3D printing an ear-fit sensor, the real time monitoring of core body

temperature can be realized with wireless connectivity.¹¹⁸ A hybrid supercapacitor with temperature sensor system using all-fiber materials was fabricated by 3D printing.¹⁰⁵ By introducing up-conversion nanoparticles in the stretchable polymer based optical fibers, wearable health monitoring systems can sustain strain up to 80 %.¹¹⁹ Inkjet printing was used for fabricating a highly sensitive temperature detector with high linearity of 0.15 % from -20 to 100 °C.¹²⁰ Monolithic laser induced reductive sintering was developed for fabricating temperature field measurement device.¹²¹ 3D printing also showed the capability for building integrated electronics devices with supercapacitors powered temperature sensors, with 1.2 % per degree sensitivity.¹²² From the above literature, it is interesting to note that there is a trend for the development of collaborative systems for temperature sensors with integrated energy storage devices, rather than a single component.

4.2.3 Glucose sensors

Digital manufacturing provides advantageous features with customized designs for glucose sensing, as shown in Figure 13. The flexible glucose sensor with inkjet printed gold nanoparticles was developed with a linear range of 0 to 40 mg/dL and a detection limit of 0.3 mg/dL.¹²³ By inkjet printing CuO nanoparticles on platinum electrodes, a non-enzymatic electrochemical sensor showed a high sensitivity of 1.6 mA/cm² per mM with a wide linear range.¹²⁴ A saliva wearable glucose monitoring device was 3D printed with a glucose detection limit of 27 μmol/L.¹²⁵ Laser scribed carbon paper can provide active sites of electronic/ionic pathways, with a glucose detection limit of 3.6 mA/mM cm².¹²⁶ Superhydrophilic patterns arrays can be fabricated via two-stage laser scribing on superhydrophilic graphene for sweat droplet manipulations and electrochemical measurements.¹²⁷ An inkjet printed organic electrochemical transistor (OECT) glucose detector with up to 30% stretchability can operate at a 1 μM sensitivity.



Figure 13 Digital manufactured wearable glucose sensor on human wrist.

4.2.4 Pulse rate sensor

The continuous monitoring of human heart rates provides essential feedbacks for evaluating the physiological condition of the elderly people. Self-powered pulse rate monitoring can be realized by laser printing of piezoelectric sensors, as shown in Figure 14.¹²⁸ Heart rate monitoring can be realized by self-powered TENG fabricated by 3D printing. A heart rate of 6

s duration can be realized by running for 10 min.¹²⁹ Stereo laser lithography could be used to build microsprings with CNT as an arterial pulse sensor within a wearable plaster.¹³⁰

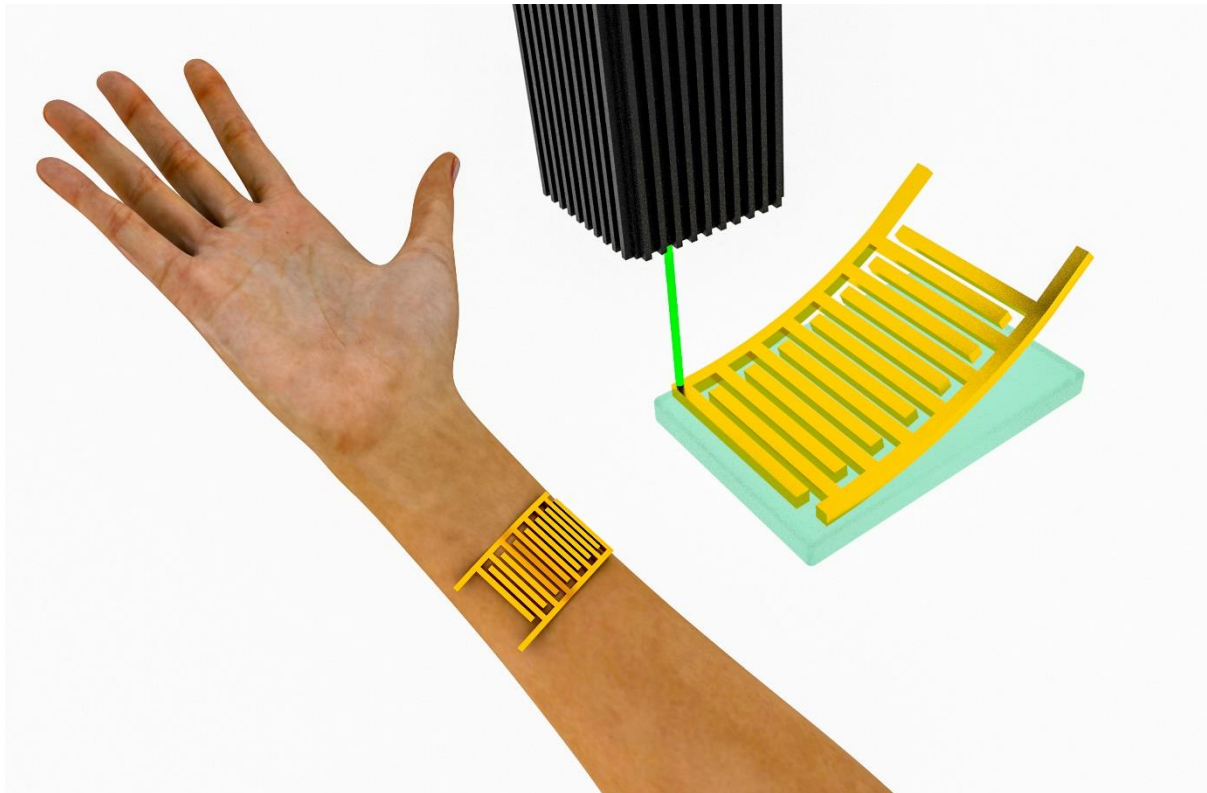


Figure 14 Pulse rate monitoring with laser patterned piezoelectric sensor.

4.2.5 Electroencephalography

Electroencephalography (EEG) is an important method for recording brain waves through skin-contact monitoring, and is a mature technique for experimental neuroscience. Digital manufacturing of electroencephalography electrodes can provide novel solutions with higher customization and sensitivity. 3D printed dry EEG electrodes with low cost and quick response make it highly customized for brain-computer interface applications.¹³¹ Direct laser writing of microneedle arrays can be realized on flexible substrates for EEG electrode applications.¹³²

4.2.6 Humidity sensor

Differing from inkjet printing or 3D printing, the laser writing method is characterized with all dry procedure, which makes it a more effective method for creating humidity sensors. Laser writing on polyimide and paper bilayers can create a dual sensor for sensing humidity and force.¹³³ Laser induced graphene can also be transferred to other substrates, such as by peeling off the PDMS capping the scribed polyimide, to fabricate a flexible humidity sensor with fast response for adsorption at 0.9 s and desorption at 4.5 s.¹³⁴ A high resolution humidity sensor with spatial resolution down to 12 μm can also be created with a shorter wavelength of 405 nm.¹³⁵

4.2.7 Gas, pH and photon sensor

Laser induced graphene can also work for gas sensing. An electronic nose can be made by transferring laser induced graphene onto a PET substrate with Pd nanoparticles loading, for sensing non-polar H_2 molecules.¹³⁶ Laser irradiation on the screen printed TiO_2 film can also work as a wearable gas sensor for ethanol detection.¹³⁷

The laser writing of PANI and carbon electrodes can create a pH sensor with linearity of 53 mV/pH in the physiological pH range of 4 to 10.¹³⁸ Inkjet printing can be used to fabricate biosensors for the selective detection of lysozyme due to the strong adhesion of a single DNA to CNT.¹³⁹

Wearable photodetectors can provide continuous measurement for UV and other optical radiation. Inkjet printing was developed for fabricating a photodetector using ZnO, with a short response time of 0.3 s and a high on/off rate up to 3,525.³⁴ Laser writing was another effective way to create a photodetector using porous rGO and ZnO, with high photocarrier generation, due to the highly porous structures.¹⁴⁰ 3D printing of polymer semiconductors also demonstrated successful fabrication of flexible photodetectors with quantum efficiency up to 25.3%.⁶¹

4.2.8 Transistors

Wearable transistors are very popular types of electronics devices due to the high-performance nature of their advanced semiconductors. The high quality and fine feature size control of the channel components provide the key for high performance transistors. Digital manufacturing has been widely studied for creating various types of flexible transistors, as illustrated in Figure 15. Inkjet printing was used for printing organic thin film transistor (TFT) arrays at 2 μm channel length with using semiconducting ink.¹⁴¹ Single wall CNTs have been successfully inkjet printed for pressure sensing, with ultralow operation voltage down to 1 V.¹⁴² The IZO and silver composite channels were fabricated using inkjet printing with a strong thermionic transport regime at a subthreshold slope as low as 16 mV/dec.¹⁴³ A fully inkjet-printed OTFT with Schottky barrier was realized with ultralow power consumption, less than 1 nW for amplifier applications.⁴⁰ The integration of dual-mode transistors were demonstrated using the combination of inkjet and extrusion printing for long-durability wearable electronics, using various emerging materials such as CNT, 2D materials, and oxide semiconductors, etc.¹⁴⁴ High performance transistors have also been fabricated with inkjet-printed CNT and polymers, operating at 1 V, with mobilities up to 30 cm^2/Vs .³⁸ At the same time, laser writing was also used for building transistor based sensing systems. Microcapillary arrays have been built using direct laser writing for accurate sweat droplet acquisition and precise sample delivery to the OECT.¹⁴⁵ Laser patterned capillary arrays could also help prompt the detection of NH_4^+ and Ca^{2+} in human sweat using the OECT.¹⁴⁶ More advanced transistors with the combination of multiple digital manufacturing methods will be developed in the near future with high performance sensing and fine control of the respiration droplets.

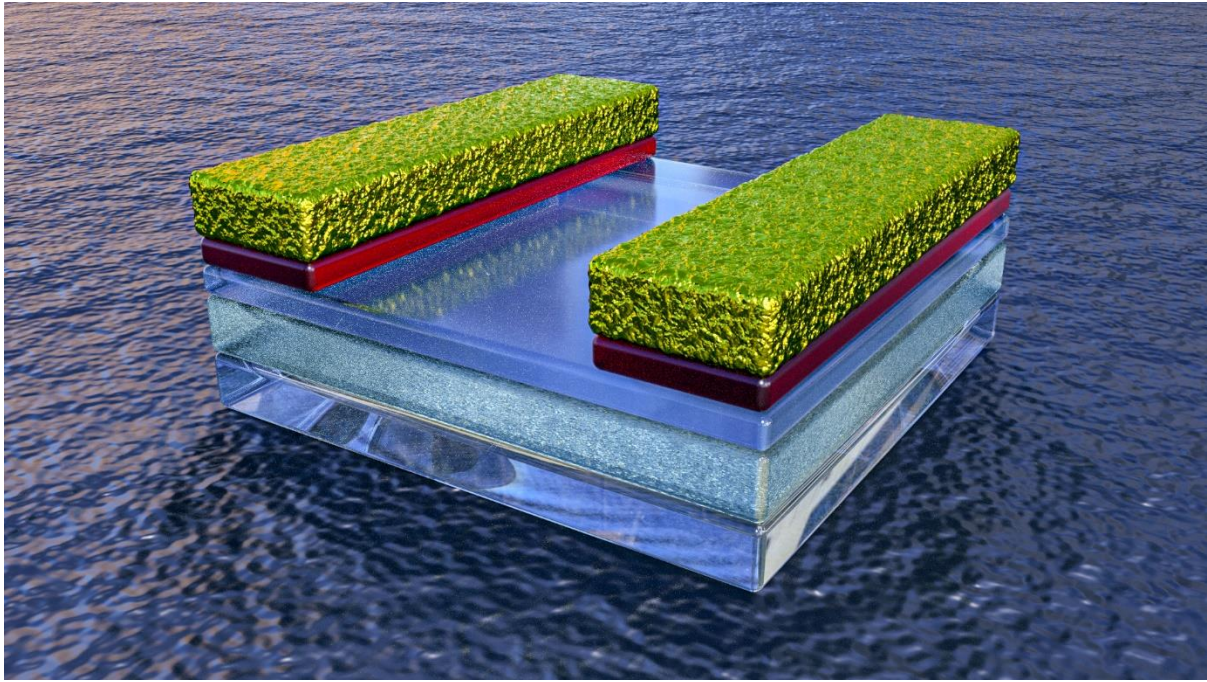


Figure 15 Illustration of multilayer transistor.

4.2.9 Smart gloves

Digital manufactured conducting materials can be used as conducting pads for electrical conduction of the power source to the electronic components. Due to its easy availability, the gloves have been widely examined as wearable flexible substrates. 3D printing of MWCNT had been used to fabricate conducting pads and strain sensors on gloves for monitoring the movement of human hands.¹⁴⁷ Thermoplastic polyurethane (TPU) can also be added to the 3D printing of MWCNT to enhance the stretchability up to 50 %.¹⁴⁸ The 3D printing of perovskite nanocomposites can be used as the piezoelectrical pressure sensor on the surface of boxing gloves for pressure mapping.¹⁴⁹ 3D printing can also be used for fabricating elastomers to hold the liquid metal for wearable gloves based electronics.¹⁵⁰ At the same time, inkjet printing was also utilized to fabricate the connecting pads on polymer gloves using Ag nanodendrite.¹⁵¹ Laser writing technologies could also create artificial patterns with laser reduced copper on the 3D printed surfaces for prosthetic applications.¹⁵²

4.2.10 E throat

Directly laser writing on polyimide has been studied for creating an actuator for converting electricity into sound energy, which could help patients with speaking difficulties.¹⁵³ The addition of PVA can enhance the sound-emitting ability at a 2 mm distance at 0.38 W.¹⁵⁴

4.2.11 Wearable heaters and personal cooling

3D printing of BN/PVA composites can result in electrical cooling in textiles for thermal management.¹⁵⁵ Flexible Bi₂Te_{2.7}Se_{0.3} nanoplate inks can be printed using 3D conformal aerosol jet printing, with outstanding power factors of 730 $\mu\text{W}/\text{m K}^2$.¹⁵⁶

Other applications for wearable electronics are also available by digital manufacturing, such as resistive switching, photobiomodulation,¹⁵⁷ smart bandaging for wound management,^{158, 159} etc.

4.3 Wearable display

As there are several reviews discussing the comprehensive developments of wearable display application,^{160, 161} herein the recent achievements are reviewed. Self-healing feature of the light-emission diodes (LEDs) was first investigated by the non-digital manufacturing methods, with the excellent performance of recovering its luminance after broken or deformation.¹⁶² Later, Tan *et al.* reported the 3D printable methods for self-healable and optoelectronic stretchable devices which boost the application of customized patterned design, with working condition under alternating voltage of 23 V and a frequency below 1 kHz, achieving a brightness of 1,460 cd m^{-2} at 2.5 V μm^{-1} .¹⁶³ The self-healing properties and the rapid development of customized digital manufacturing provide a promising solution for applying in the smart phones. Additionally, digital manufacturing shows satisfying ability for dealing with organic or inorganic perovskite material. Chao's group has figured out three protective encapsulation materials for perovskite nanocrystals which enables the 3D printing production and maintains the excellent optical properties of the perovskite materials.¹⁶⁴ And the perovskite nanocrystal-PCL composite films were incorporated to build a white light-

emitting diode with a CIE coordinate (0.33, 0.33). The possibility of laser writing 3D patterns of perovskite quantum dots was also proved by Dong's research group.¹⁶⁵

5. Future Perspectives

Although existing digital manufactured functional materials have been developed for many aspects of wearable electronics, there is plenty of room for future development. With the emerging developing of novel functional materials, higher performance materials have been frequently reported.^{166, 167} Semiconducting materials with higher mobility and flexibility have been developed, which can further improve the performance of the wearable electronics.¹⁶⁸ Metallic and alloy nanoparticles with active sites are emerging, which can lead to better selectivity and higher sensitivity toward molecule sensing.¹⁶⁹ The study of these novel materials for digital manufacturing is vital for constructing future wearable electronic components.¹⁷⁰

At the same time, some novel digital manufacturing methods are being developed for building higher accuracy devices in a shorter time. Previous 3D printing and inkjet printing were relatively slow in fabricating large numbers of products.¹⁷¹ The developing of novel 3D printing methods, such as the digital projection method, can significantly reduce the production time for printing various devices.¹⁷² However, incorporation of functional materials into these novel methods is lacking, and needs many future experimental studies and optimization. The inkjet printing speeds can also be boosted by introducing more nozzles in the printing head, which needs better pixelization optimization for printing more droplets simultaneously.¹⁷³

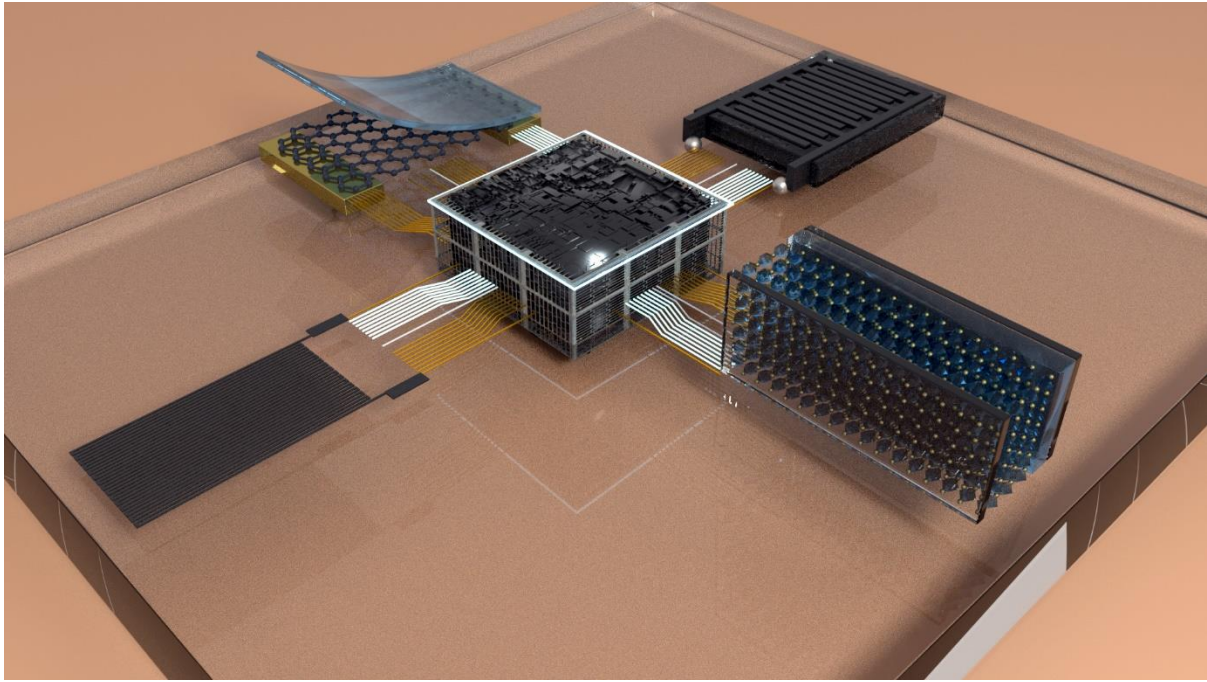


Figure 16 Illustration diagram of digital manufactured future integrated wearable systems, with solar cell, battery, transistor, and strain sensor, connected to microcontroller.

Furthermore, the integration degree of wearable electronics can be improved, as shown in Figure 16. Power generating devices, including solar cells and TENGs, can be better integrated with energy storage devices, including batteries and supercapacitors, as self-powered frameworks for long term power systems. Wireless transmitted antennas can be integrated with various wearable sensors for synchronizing the signals to smart phones.¹⁷⁴ The sensor networks can generate a large amount of data, such as body temperatures, sweating rates and corresponding components, which can be transferred wirelessly to smart phones and further analyzed with artificial intelligence for systematically monitoring of the human physiological states.^{175, 176} At the same time, the anti-bacterial properties of the functional materials can provide additional advantages for the wearable electronics. The superhydrophobic and photothermal effect of the semiconducting and plasmonic nanoparticles can offer anti-bacterial properties for long-term wearable devices.¹⁷⁷ Lastly, the privacy of the collected data should be handled properly at a safe level, without the leakage of personal data to third parties.^{178, 179}

6. Conclusions

In this paper, the state of the art of digital manufacturing of the functional materials for wearable electronics is reviewed. Inkjet printing, 3D printing and laser writing are the essential methods for creating artificially designed devices for health monitoring and usages. The emerging development of functional materials provides more choices for building high performance, flexible and stretchable devices. The integration of the power system, wireless transmission and various sensors can provide long-term and systematically daily monitoring. These emerging and interdisciplinary aspects are worthy of further study for future developments in the fields of functional materials and novel manufacturing methods, for building more comprehensive devices for the characterization of physiological information.

Acknowledgments

J. Lin and Z. Zhu contributed equally. G. Li acknowledges the funding support by State Key Laboratories in Hong Kong from the Innovation and Technology Commission (ITF) (No. 1-BBX9) of the Government of the Hong Kong Special administrative Region (HKSAR), China.

The authors declare no conflict of interest.

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