3D printing of multilayered and multimaterial electronics : a review

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Title: 3D printing of Multi-Layered and Multi-Materials Electronics: A Review

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Abstract

3D printing, also known as additive manufacturing, is a manufacturing process in which the materials are deposited layer-by-layer in an additive manner. With the advancement in materials and manufacturing technology, 3D printing has found its applications in the field of electronics manufacturing. Initially, 3D printing is used for the fabrication of electronic components with single material designs such as resistors, inductors, circuits, antennas, strain gauges, etc. Recently, there are many works involving the use of 3D printing fabrication techniques for advanced electronic components and devices such as parallel plate capacitors, inductors, OLED, photovoltaic, transistors, displays, etc. which involved multi-layer multi-material printing. Despite

these many works, there has been no review on design and fabrication consideration for the 3D printing of multi-layered and multi-material (MLMM) electronics. As such, this review aims to summarize the current landscape of 3D printing of MLMM electronics and provide some insights on the design consideration, fabrication strategies, and challenges of 3D printing of MLMM electronics. In particular, the focus will be laid on discussing the interface between different materials such as surface wettability, surface roughness, material compatibility, and the considerations for post-processing treatments.

1. Introduction

1.1 Overview of manufacturing approaches for multi-layered and multi-material electronics

Figure 1. The comparison between traditional subtractive approach and the modern additive manufacturing approach [Reprinted with permission from ref^[1]. Copyright 2017 Royal Society of Chemistry].

Upcoming advanced electronic components and devices are receiving widespread attention as a type of innovative technology that will drastically change our daily lives. As the multi-layered and multi-material (MLMM) fabrication process will make it possible to speed up their realization, there has been increasing interest in using MLMM fabrication technology for producing various high-performance and flexible electronics, such as transistors,^[2-4] energy storage devices,^[5-8] low-cost sensors,^[9-11] radio frequency identification (RFID) tags^[12-14] and other disposable electronic

devices^[15-18] in recent years. MLMM electronics currently can be fabricated using two main approaches: the traditional subtractive approach and the modern additive manufacturing approach.^[19] As shown in Figure 1, the manufacturing process of RFID antennas is adopted as an instance to illustrate the difference between traditional subtractive approach and the modern additive manufacturing approach. As the traditional fabrication methods for MLMM electronics consist of a series of subtractive processes, such as etching and film releasing, which brings forth various disadvantages including hazardous chemical preparation,^[20] complicated and expensive set-up, and a considerable amount of acid-base material wastage. On the contrary, the additive manufacturing approach is considered as one of the most promising approaches due to its clear advantages in terms of reduction in chemical wastage, time bottleneck, and setup cost, which will bring about the possibility of MLMM electronics in a much simpler, faster and cost-effective way. As shown in **Table 1**, the modern additive manufacturing approach for producing MLMM electronics consists of two main types: contact printing and non-contact 3d printing. Unlike traditional fabrication methods, additive manufacturing approach has the capability of fabricating MLMM electronics on a wider range of flexible substrates, such as fabrics, papers, and polymer films,^[21, 22] which provides a promising methodology to manufacture novel MLMM functional electronic devices. Furthermore, the integration of different additive fabrication methods such as hybrid printing is beneficial to the fabrication of MLMM electronics in complex structures.^[23] Therefore, the additive manufacturing approach is currently the most receivable technology for the fabrication of advanced MLMM electronics, and the remaining section will systematically review the working principles of different additive manufacturing methods with advantages and disadvantages.

Table 1. Main additive manufacturing techniques for producing electronics

Main types	Fabrication process	Fabrication techniques	Material types	References
Gravure printing	Contact printing	Roll-to-Roll printing	Ink-based	[24-28]
Flexographic printing	Contact printing	Roll-to-Roll printing	Ink-based	[29-32]
Offset printing	Contact printing	Roll-to-Roll printing	Ink-based	[33-37]
Rotary screen printing	Contact printing	Roll-to-Roll printing	Ink-based	[8, 18, 38-40]
Flatbed screen printing	Contact printing	Sheet based printing	Ink-based	[41-43]
Fused deposition modeling	Non-contact printing	3D printing	Filament-based	[44-47]
Direct ink writing	Non-contact printing	3D printing	Filament-based	[48, 49]
Organic vapor jet printing	Non-contact printing	Vapor deposition	Vapor-based	[50-54]

Laser-induced forward transfer	Non-contact printing	Laser direct write	Vapor-based	[55-58]
Aerosol jet printing	Non-contact ink writing	3D printing	Ink-based	[59-64]
Inkjet printing	Non-contact ink writing	3D printing	Ink-based	[65-68]
Hybrid printing	Non-contact printing	3D printing	Multi-material based	[69-74]

1.2 Contact printing techniques for MLMM electronics

Contact printing is a conventional additive-based electronic fabrication method that requires the printing pad to be in contact with the target substrate to transfer the designed inked patterns. As shown in **Figure 2**, the contact printing processes typically employ an engraved roll or a single sheet to transfer the functional ink onto the target substrate. However, the pros and cons of different contact printing techniques may vary with the specific printing principles, thus it is important to compare the working principles of each contact printing technique systematically.

As shown in Figure 2(a-d), roll-to-roll (R2R) printing techniques which include gravure printing, rotary screen printing, flexographic printing, and offset printing have been widely adopted as representative contact printing approaches for printing MLMM electronics. For gravure printing, the printing process consists of two phases. In the first phase, a gravure roller collects inks from the ink fountain, and a blade is used to control the volumes of transferred ink. Following that, the collected ink can be transferred to the substrate by adjusting the pressure between the impression roller and the gravure roller. Similarly, as an ink-blocking stencil can block unnecessary areas on the substrate from coming in contact with the functional ink, rotary screen printing can print the designed patterns on the substrate by the imposed pressure from the impression roller. Unlike gravure printing and rotary screen printing, flexographic printing (or offset printing) adopts a soft plate roller (or blanket roller) as a medium substrate to transfer the controlled amounts of ink, the patterns will be printed indirectly onto the substrate. On the other hand, flatbed screen printing, which is another variant of screen printing technique, is also receiving great interest in recent years. **Figure 2(e)** illustrates the manufacturing process of flatbed screen printing, and the comparison between different contact printing techniques is shown in Table 2.

Figure 2. The comparison between different contact printing techniques [Reprinted with permission from ref^[1]. Copyright 2017 Royal Society of Chemistry]. (a-d) Roll-to-roll printing. (e) Flatbed screen printing.

Table 2. Technical specifications of classical contact direct printing techniques

Printing techniques	Advantages	Disadvantages
Gravure printing ^[75, 76]	<ul style="list-style-type: none"> - High throughput - Suitable for substrates with varying flexibility - Capable of using low-viscosity inks - Capable of printing different film thicknesses 	<ul style="list-style-type: none"> - Mask is indispensable - High setup cost - Limited to flat surfaces - The bottom layer may be damaged
Flexographic printing ^[77-79]	<ul style="list-style-type: none"> - Suitable for compressible substrates - Suitable for water-based or oil-based ink - Fast printing speed 	<ul style="list-style-type: none"> - Mask is indispensable - Relatively low print resolution - The bottom layer may be damaged
Offset printing ^[80-82]	<ul style="list-style-type: none"> - High productivity - High printing quality - Capable of using low-viscosity inks - Flexible printing process 	<ul style="list-style-type: none"> - High setup cost - A medium substrate is required - The bottom layer may be damaged
Screen printing ^[83-86]	<ul style="list-style-type: none"> - Suitable for numerous substrates - Low-cost mass production - Fast printing speed - Repeatable printing accuracy - Suitable for numerous substrates 	<ul style="list-style-type: none"> - A medium substrate is required - Post-processing is required - Need highly viscous ink - Need controlled printing condition - An ink-blocking stencil is required

1.3 Non-contact 3D printing techniques for MLMM electronics

Despite the many advantages, the use of a contact printing technique for fabricating MLMM electronics is still limited due to printing quality and setup cost. Non-contact 3D printing is an

emerging non-contact printing technology for producing flexible and low-cost MLMM device applications. Unlike contact printing, non-contact 3d printing technology is maskless, and able to achieve higher resolution and produce less waste, and therefore it has attracted great attention in various fields.^[87-89] As shown in **Figure 3**, the 3D printing techniques for producing MLMM electronics can be classified into three main types: (i) material extrusion-based printing such as fused deposition modeling (FDM) and direct ink writing (DIW), (ii) vapor-based printing such as laser-induced forward transfer (LIFT), and organic vapor jet printing (OVJP), as well as (iii) material jetting-based printing such as inkjet printing (IJP) and aerosol jet printing (AJP).

Figure 3. Working principles of 3d printing techniques for printed electronics (a) Fused deposition modeling [Reprinted with permission from ref^[90]. Copyright 2018 MDPI]. (b) Direct ink writing [Reprinted with permission from ref^[48]. Copyright 2020 Wiley]. (c) Laser-induced forward transfer. (d) Organic vapor jet printing. (e) Continuous inkjet printing, (f) Drop-on-demand inkjet printing, (g) Aerosol jet printing [Reprinted with permission from ref^[60]. Copyright 2019 Springer].

1.3.1 Material extrusion-based printing

FDM is also known as fused filament fabrication or filament freeform fabrication, which is a material extrusion additive manufacturing technology that is based on continuous filament of a thermoplastic material. **Figure 3(a)** shows the working principles of the FDM technique. During the printing process, the thermoplastic filament is first fed from a large spool and melted by the heated printer extruder head. After that, the melted filament is extruded by mechanical pressure

from rollers and printed onto the substrate. Despite FDM has been adopted for fabricating MLMM electronics, the electrical performance of printed electronics is limited due to the printing quality and resolution. Therefore, it is important to optimize the flow geometry of the extruder, heating method, and the melt flow behavior of a non-Newtonian fluid to improve the printed line quality.

Direct ink writing (DIW) is very similar to the FDM printing technique except that the printing material used in DIW is normally functional inks instead of filaments (**Figure 3b**). For DIW, the functional ink that is first stored in the syringe barrel will then be extruded out from a nozzle and get deposited on a substrate.^[91] The functional inks for DIW can be in the form of low viscosity ink or shear-thinning ink. Low viscosity ink flows easily through the nozzle, whereas shear-thinning ink requires a much larger driving force. When using low viscosity ink, a curing system will have to be used to ensure that the extruded filament cures instantly to retain its 3D structure. As such, the functional inks tend to have additional binding material mixed with the functional material which can lower the electrical performance of the functional material.^[92] In general, there are different types of dispensing methods, namely pneumatic, piston and screw. It is vital to use proper parameters and environments for printing to ensure the printed filaments form a stable 3D structure.

1.3.2 Vapor-based printing processes

As shown in **Figure 3(c)**, laser-induced forward transfer (LIFT) is a representative method of laser direct-writing (LDW) and has been employed for the deposition of a variety of materials for electronic applications. In this technique, a high-energy focused laser pulse has been used to supply sufficient energy to heat the interface of the donor thin film. Due to energy accumulation, the melted interface of the solid film can be converted into vapor metal and transferred onto a target substrate to produce high-quality electronic patterns. In contrast to LIFT, for an OVJP printing system as shown in **Figure 3(d)**, the cells containing the molecular organic source material are heated and the generated saturated source vapors are transferred by a hot inert carrier gas. The gas mixture then expands through a microscopic nozzle and impacts the substrate with high speed. As the organic vapors will condense on the freeze substrate while carrier gas will disperse after the impact, a line with high resolution and high density is fabricated on the substrate.

1.3.3 Material jetting-based printing processes

Due to the advantages of higher resolution and less waste, there has been increasing interest in using droplet-jetting-based direct ink writing technologies, especially IJP and AJP for producing MLMM electronics. As shown in **Figure 3(e-f)**, IJP can be classified into continuous inkjet (CIJ) printing system and drop-on-demand (DoD) printing system based on different working principles. For a CIJ printing system, due to the influence of an electric field, the charged continuous droplets will be captured for reuse while the uncharged droplets will print onto a substrate as printed patterns. Unlike CIJ printing, a DoD printing system will generate and eject droplets onto a substrate directly due to the applied momentum. Compared with IJP, AJP is an emerging non-contact ink writing technology for fabricating MLMM electronics (**Figure 3g**). In this technique, the functional ink is first atomized into droplets (2 to 5 μm) and transferred into the print head by a carrier gas flow. Following that, as a sheath gas flow will enwrap the ink aerosol in the nozzle cylindrically, the generated droplets will exit the nozzle tip and impact the moving substrate with high speed, and a line with high density is produced on a substrate. AJP has flexible working height, and is capable of printing high viscosity ink over nonplanar substrates with higher resolution and without clogging.

1.3.4 Comparison between classical 3d printing technologies

The comparison between classical 3d printing technologies for fabricating MLMM electronics is shown in **Table 3**. Due to the low printing resolution, the filament-based FDM is not suitable for printing electronic devices which require fine feature size. Despite the vapor-based OVJP and LDW is capable of printing with high resolution, the throughput is limited due to relatively lower print speed. On the other hand, as the ink-based IJP and AJP are capable of printing on flexible substrates with higher resolution, they are currently the most receivable 3d printing techniques for producing MLMM electronics and will be systematically discussed in the next.

Table 3. Technical specifications of classical non-contact direct printing techniques^[1, 45, 46, 49, 50, 60, 92-95]

Printing methods	Ink viscosity (cP)	Feature size (μm)	Process Speed (m/min)	Advantages	Disadvantages
FDM	/	200	1-10	<ul style="list-style-type: none"> - Print with multi-materials - Relatively high print speed - Without coffee-ring effect - Low setup cost 	<ul style="list-style-type: none"> - Low resolution - Nozzle clogging - Low printed line quality - Process instability

DIW	10^2 - 10^8	Depending on nozzle size	Up to 3	-Able to fabricate multi-material -Can achieve free-standing structure	-Requires ink with binder that can result in lower electrical property
LIFT	/	<20	Depending on laser's speed	- No requirement of vacuum - Multi-materials printing - Produce less waste	- Limited to planar surface - Laser may damage pattern - High-power laser consumption
OVJP	/	<25	$> 4 \times 10^{-6}$	- No requirement of solvent - Multi-materials printing - Produce less waste	- Limited to planar surface - Low process speed - Supplying system is required
IJP	10-20	>30	15-500	- Suitable for various substrates - Fabricate with multi-materials - Produce less waste - Low contamination	- Nozzle clogging - Coffee-ring effect - Limited to planar surface - Limited ink formulation
AJP	1-2500	10-200	Up to 12	- Suitable for various substrates - Print on non-planar surface - High resolution - Less clogging	- Supplying system is required - Limited ink formulation - Coffee-ring effect

1.4 Analysis of ink-based 3d printing process for printed electronics

As shown in **Figure 4**, the basic fabrication process of an ink-based 3d printing technique such as IJP and AJP consists of three main steps: (1) pre-printing, (2) intermediate-printing, and (3) post-printing. For the pre-printing stage, as the ink properties and substrate properties have a great impact on ink printability, it is important to choose suitable ink and substrate to improve the ink printability, otherwise, the conductive line may be printed discontinuously or with high overspray. During the intermediate-printing stage, due to the influence of interfacial interaction, inter-layer compatibility, and the complex interaction between different process parameters, the conductive line may be printed with high edge roughness and insufficient thickness. Therefore, there is a need to optimize the intermediate printing process to ensure the controllability of printed line morphology. During the post-printing stage, the printed electronic devices will be post-processed by an oven, laser, or UV light to improve electrical conductivity. However, as the post-printing process will cause non-uniform distribution of nanoparticles around the cross-sectional area, the lines are printed inhomogeneous and prevented from being widely applied to various electronic components. Based on the above discussion, it can be seen that the three main steps of the basic

fabrication process have a great impact on the printed line quality, which will significantly affect the electrical performance of the produced electronics. **Table 4** lists the main influencing factors of the three main steps. More in-depth discussions on each printing and sintering stage will be discussed in the following sections.

Figure 4. Analysis of ink-based 3d printing process and the influence on MLMM electronics

Table 4. Main influencing factors of non-contact direct ink writing techniques

Categories	Main influencing factors
Pre-printing factors ^[96-99]	Ink properties: ink concentration, ink viscosity, particle size, surface tension Substrate properties: surface wettability, contact angle, surface roughness
Process parameters ^[49, 100-109]	DIW: print speed, nozzle size, pressure IJP: droplet spacing, jetting delay, voltage, waveform, nozzle size, printing height AJP: SHGFR, CGFR, print speed, tip size, atomizer current, working height
Post-printing factors ^[110-114]	Oven: sintering time, temperature, sintering cycles Laser: laser power, scanning time, speed UV: UV spectrum, UV power, scanning speed, time

2. Design & fabrications considerations, approaches, and strategies for 3D printed MLMM electronics

The design and fabrication processes for MLMM electronics is very complicated as it involves sequential printing and treatment of different materials at various printing and treatment conditions. As such, there more factors that need to be taken into considerations for 3D printing of MLMM electronics. This section will discuss the design & fabrication considerations, approaches, and strategies for 3D printed MLMM electronics in the following categories, namely single material multi-layer printing, co-planar multi-multi-material printing, and multi-layered multi-material printing as depicted in **figure 5**. The discussion begins with the overprinting of single materials on a substrate which is the basis of MLMM electronics. Following that, the printing of multiple materials on a substrate (co-planar design) will be discussed. Lastly, the printing MLMM electronics involving the stacking of a material on a different material will be elucidated.

Figure 5. Schematic showing different constructs of MLMM electronics, namely (a) overprinting of a single material, (b) coplanar multi-material printing, and (c) multi-layered and multi-material printing of electronics.

2.1 Overprinting of a single material

Single material printing of functional inks involves the deposition of the printing materials on a substrate material directly. It is the foundation of 3D printing of MLMM since it creates the first functional layer of an MLMM electronic component or device. As the fabrication process involves only a substrate material and an ink material, the main factors that can affect the quality of the printed film are either the rheological property of the ink and the ink-substrate interaction depending on the printing technology. In particular for DIW techniques, the homogeneity and the rheological property of the inks play a critical role in ensuring the uniformity and stability of the printed filaments, thus the printed structures.^[115] Therefore, the functional inks for DIW are

normally pre-treated to remove air bubbles that can cause discontinuity of the extruded filament.^[116, 117] As for material jetting-based 3D printing techniques such as IJP and AJP, the ink-substrate interaction plays a significant role in determining print quality. The ink-substrate interaction can often be indicated by the surface wettability (**Figure 6a**). Surface wettability is an important consideration when using liquid-based functional ink because it has a strong influence on the adhesion strength of the printed material to the substrate and the printability of the ink for high-resolution printing. Typically, surface wettability is determined by the surface tension of the ink, and the surface energy, morphology, and roughness of the substrate.^[118] Good surface wettability can often be achieved by using inks with low surface tension and smooth substrates with high surface energy.^[119] With proper optimization of printing parameters, good surface wettability ensures the formation of continuous conductive tracks or printed patterns. In contrast, poor surface wettability often leads to poor drop stability on the substrate, resulting in the breakup of the deposited liquid structure due to dewetting, hence undesirable print quality.^[119] Besides, poor surface wettability also leads to poor adhesion strength, making the printed film peel off easily from the substrate.^[120] The surface wettability can be improved by using various surface treatments such as UVC treatment^[121-123] and oxygen plasma treatment.^[124, 125]

For solution-processed ink, the surface morphology of the substrate has a significant impact on the final deposition pattern of the printed material. As the solvent of the ink requires time to evaporate, small microstructural features such as pores and grooves on the rough substrate (e.g., parts fabricated using fused deposition modeling and selective laser sintering) can cause undesirable spreading of ink due to capillary action.^[126, 127] This can lead to geometrical distortion of printed patterns or discontinuity within the printed pattern, resulting in inconsistent performance or functionality (**Figure 6a**).^[128] For the fabrication of small line pitch spacing components such as interdigitated electrodes, the undesirable spreading ink could bring about short-circuiting issue. The severity of the effect depends on several factors such as the rheological property of the ink, distribution of the active material in the ink, and the surface roughness of the substrate. Inks with higher viscosity can be used as the viscous force overpowers the surface tension that causes the capillary action.^[126] However, the limiting factor, in this case, is the maximum allowable viscosity processible by the printing techniques. Alternatively, it is suggested that the rough surface be smoothed by either polishing or coating.^[129]

Other than the printability of the ink and the adhesion of the printed material, the homogeneity of the printed materials also plays an important part in dictating the performance of the printed device (**Figure 6b**). The homogeneity of the printed material is determined by the distribution of functional materials on the substrate (e.g., nanoparticles or organic macromolecules) as a result of the rheological property and evaporation mode of the ink, and interactions between the particle, hydrodynamic forces, and the substrate.^[130, 131] In most cases, a homogeneous film structure is preferred as it results in a more consistent and reproducible performance of electronic components or devices. Homogenous film structure can be achieved by using particles that have strong interaction with the substrate, thereby suppressing the influence of hydrodynamic flow (e.g., capillary flow) and surface tension on the motion of nanoparticles during the evaporation process.^[130-133]

The performance of an electronic component is normally determined by the conductivity of the conductive materials or the dielectric strength of the dielectric material.^[134] It is often desirable to have conductive materials with high electrical conductance (or low electrical resistance) as it can result in electrical components with lower energy loss.^[135] The electrical resistance of the printed film is dictated by various factors such as the type of material (type of inks), the sintering treatment, and the geometry of the printed film. For a selected type of conductive ink and sintering treatment, the electrical resistance of printed patterns with a fixed footprint can be adjusted by changing the film thickness. For dielectric materials, the dielectric strength is often dictated by its film thickness and the material homogeneity on the substrate.^[135] In order to achieve high film thickness, overprinting technique is often used to deposit the same material multiple times (multi-layer printing of a single material) to additional layers of material. In order to ensure good deposition of ink and the formation of homogeneous film, several fabrication considerations need to be taken care of. Firstly, it is important to ensure that the deposited ink has completely dried before printing the next layer. Otherwise, there is a possibility of the solvent-rich incoming ink drops to mix with the solvent-depleted deposited ink, resulting in a change in concentration of the deposited ink which could yield non-uniform film. This can be eliminated by heating the substrate to accelerate the evaporation process of the solvent. Secondly, the substance within the dried films that have not undergone sintering or curing process is normally loosely bonded to the substrate. Thus, depositing solvent-rich ink onto the uncured dried film could cause the substance from the dried film to redissolve in the deposited ink, thereby affecting the homogeneity of the deposited film. This is

phenomenon is more obvious for aerosol jet printing where the interaction between the strong impinging flow and solvent-rich ink could disrupt the homogeneity of the uncured dried film (**Figure 6c**).^[136-138] This can be easily overcome by integrating a sintering or curing system into the printer to allow in-situ treatment of the dried film before printing the next layer. Lastly, the sintering and curing method for treating the thick dried film has to be carefully selected. It should be noted that certain sintering techniques such as photonic and laser sintering have limited penetration depth which could result in incomplete sintering or curing of the thick film (**Figure 6d**).^[139] Also, the dried film may still contain some amount of unevaporated solvent trapped within the thick film.^[140, 141] In such cases, rapid sintering may cause rapid expansion of the solvent which may rupture the film (**Figure 6e**), resulting in detrimental effects on the performance of the printed film. Also, the cracks and film rupture can also affect the printability of the subsequent layers for the case of MLMM electronics printing. Therefore, it is recommended to slowly ramp the sintering temperature to avoid film rupturing.

Figure 6. Design and fabrication considerations for single material printing, namely (a) surface wettability and surface roughness, (b) homogeneity of film, (c) process-induced anomalies, and the limitations of post-processing technique for thick film such as (d) limited penetration depth, and (e) surface cracks and film rupture. Reprinted with permission from ^[63]. Copyright (2019) American Chemical Society

2.2 Coplanar multi-material printing

The fabrication of coplanar multi-material printing involves the deposition of inks with different material types on to the same substrate. A prominent example of coplanar multi-material printing is the fabrication of 3-electrode electrochemical electrode systems.^[142, 143] Typically, each electrode is made up of different materials and printed separately onto the substrate. The design and fabrication considerations for such coplanar multi-material printing encompass the abovementioned for the single-material multi-layered printing. In particular, the inks may have different surface tensions depending on the solvent and additive used, thus giving different surface wettability. Also, the molecules or particles may have different surface charges and will interact differently with the substrate, thus possibly producing different film morphologies. As such, optimization of the printing process parameters for each ink type ought to be done before executing the coplanar multi-material printing as depicted in **figure 7a**.

As most inks require sintering or curing treatment, it is imperative to ensure that the deposited films of different material types can be processed within the same sintering conditions without compromising the performance of each printed material. For instance, PEDOT:PSS will suffer chemical degradation at a temperature higher than 120 °C,^[144] however gold nanoparticle requires a sintering temperature of at least 300 °C,^[145] thus such material combination should be avoided. This can be overcome by several ways, namely (1) choosing compatible materials, (2) selective sintering of printed film, and (3) sequential printing and sintering of different materials. First, it is always recommended to choose a material combination where all the selected materials have similar thermal stability or requirement on sintering condition. If getting the right material combination is impossible, selective sintering methods such as laser sintering can be used as an alternative to allow for the use of optimum sintering conditions for each material type. Otherwise, careful planning of the printing and post-processing treatment sequence for each material should be adopted. However, the fabrication process will be tedious as it requires intermediate printing

and sintering or curing for each material (**Figure 7b**). In this case, the material that has a higher requirement for sintering conditions or higher thermal stability should be printed and treated first, followed by the material that has the next lower requirement. For systems without an in-situ sintering system, alignment of the substrate or samples can be a potential issue as the samples have to be taken out from the build platform each time for the sintering or curing treatment. As such, fiducial marks should be added to the substrate for alignment purposes. Alternatively, it is recommended to use sintering-free inks that can be post-processed using freeze-drying method to prevent thermal degradation due to excessive heat.^[146] Freeze-drying post-processing technique is frequently used for treating DIW-printed electrodes to retain their shape as the process is found to have minimal impact on the microstructural arrangement nanomaterials.

Figure 7. (a) Schematic showing a possible workflow for printing coplanar multi-material printing for materials [Reprinted with permission from ref. ^[147] Copyright 2017 Elsevier]., (b) and printing and sintering strategy for materials with distinct thermal stability using intermediate printing and sintering treatments. (T_1 and T_2 represents two different temperatures with $T_1 > T_2$)

2.3 Multi-layered multi-material printing

The fabrication of multi-layered multi-material electronics components and devices involves the deposition of a material on a previously deposited film made of a different material. Thus, the

design and fabrication considerations include the aforementioned for overprinting of a single material and coplanar multi-material printing. LEDs and transistors are some of the examples of electronics components that require MLMM printing.^[50, 148] As MLMM printing is normally a bottom-to-top fabrication approach, the printing order of the material printing must be strictly followed from the bottom to the top, unlike co-planar multi-material printing which the sequence of material printing can be changed in certain circumstances. As such, it creates more constraints on the fabrication, leading to stricter requirements for the selection of the materials and the post-processing treatments.

For MLMM printing, it is essential to ensure that all the materials have similar thermal stability if thermal sintering is used. This is because selective sintering of a printed film is almost impossible for thermal sintering technique as the previously deposited materials will experience the same sintering condition. Sintering methods such as photonic sintering or laser sintering with localized heating capability could be viable treatments for reducing the degradation of previously deposited materials. Alternatively, intermediate printing and sintering/curing can be adopted for MLMM printing if the materials for the upper layers can be treated with a sintering/curing technique such as UV treatment or chemical treatment which are less damaging to the underlying materials.

Another important aspect of MLMM printing is the chemical compatibility of the inks with the previous layer, especially when the print-all-layers-sinter-together method is adopted.^[149] Interlayer mixing of materials could happen if miscible solvents are used.^[150] Therefore, selected inks for each layer must be using orthogonal solvent systems to avoid disruption of the morphology of the underlying layers.^[151] However, the process to find good and orthogonal solvents is not easy because it is normally done by trial-and-error methods in most instances. It is important to note that inappropriate use of solvent could result in swelling and an increase in surface roughness of the underlying layer, leading to a decrease in performance of the printed MLMM components.^[152]

The surface wettability of the inks with their corresponding underlying layer is paramount for ensuring good deposition of each layer of material. Unlike the coplanar multi-material printing in which the inks are deposited on the same substrate or its underlying layer, the evaluation of surface wettability of the ink for MLMM printing should be done with the corresponding underlying layer according to the printing sequence of the materials. The surface wettability of the ink at the upper layers must be checked against both the underlying layer and the substrate if the printing pattern

involves both regions.^[134] The same workflow as proposed in **figure 7a** can also be adopted for MLMM printing. In addition, intermediate treatment can affect the surface morphology of the printed layers, and hence the surface wettability.^[153] Interestingly, accurate deposition of a certain material can be achieved by controlling the surface wettability selectively. Kwon et al. demonstrated the patterning of hydrophobic material outlines to prevent the distortion of deposition pattern of the semiconducting ink due to spreading and thereby precisely defining the area for the ink.^[154]

For MLMM printing, there are two considerations related to the surface morphology, namely (1) film uniformity and (2) stair-step effect (**Figure 8a**). This is because the short-circuiting between the top and bottom electrodes would happen if the intermediate dielectric or the semiconducting layer is not able to completely cover the bottom electrodes. As such, it is critical to obtain a homogeneous film with low surface irregularity and features such as the coffee ring effect that can give non-uniform film thickness should be avoided. However, the formation of stair-step effect in MLMM printing is inevitable due to the finite thickness of each layer. In cases where films are highly ununiform with a significant stair-step effect, a thick intermediate layer should be applied to ensure complete coverage of the underlying features. For the case of intermediate printing and sintering/curing for MLMM printing, the influence of the sintering techniques on the film morphology should be considered.^[50] For example, the laser sintering technique can change the surface morphology of the printed film and photonic sintering can cause film rupture, which could affect the uniformity of the film morphology.^[140, 155, 156]

Figure 8. Design and fabrication considerations for MLMM electronics, namely (a) the effect of layer thickness of secondary layer on non-uniform surface, and (b) crack formation due to mismatch in shrinkage of materials in different layers. [Reprinted with permission from ref^[157]. Copyright 2017 IOP publishing].

Besides, other considerations for MLMM printing would be the expansion and shrinkage of each material as a result of the post-processing treatments (**Figure 8b**).^[157] In most cases, conducting layers are made up of metal and dielectric layers are made up of polymeric material, which the mismatch between the coefficient expansion is typically significant. This can result in high residual stress in the film which can lead to crack formation.^[153] Gokhale et al. have observed the formation of cracks in the silver layer covered a layer of dielectric materials sintered with intense pulsed light sintering method. The formation of cracks can be controlled by the selection of substrate and dielectric material, fine-tuning the morphology of the silver layer, and appropriate sintering parameters. In cases where the silver is partially covered by a dielectric layer, cracks can be observed on the silver layer at the edge of the dielectric layer as the dielectric layer can act as a stress concentrator that induces cracks during the sintering process.

2.4 Realizing 3-dimensional MLMM electronics

The discussion in the earlier sections mostly revolves around the interaction of the functional inks with the underlying layers for direct write-based 3D printing technology. The fabrication of

electronics using direct-write (DW) technology such as inkjet printing and aerosol jet printing normally uses inks with a functional material dissolved in a solvent. Generally, the material loading of the ink is kept low to ensure that the functional ink meets the viscosity requirement of the respective printing processes.^[158] Due to this inherent nature of the solvent-based functional inks, the use of such inks with DW technology typically produces low layer thickness due to the high-volume loss as a result of the solvent evaporation during the drying process. Consequently, 3D printing of electronics using DW technology normally results in geometrically flat components and devices, or more commonly categorized as 2D or 2.5D thin-film electronics.

3D MLMM electronics can be achieved in three ways, namely via (1) hybrid printing technology,^[127] (2) conformal printing,^[159] and (3) alternative 3D printing techniques such as FDM and DIW with conductive nanocomposite materials.^[160] Hybrid printing technology involves the integration of various 3D printing techniques to achieve 3D structural electronics. Typically, hybrid printing is done by pairing a DW printhead with a 3D printer for the conductive layers and the insulating structural material, respectively. Hoerber et al. developed a hybrid 3D printer incorporating powder bed fusion technology and aerosol jet printing technology that is capable of fabricating entirely 3D printed structures with MLMM circuitry.^[127] They also discussed the approaches and opportunities for realizing electronic functionalization of additively manufactured 3D parts such as the embedding of conductive tracks and electronic components, printing of 3D circuit tracks, surface mounting of electronic components, and printing of MLMM electronic components (**Figure 9a**). Through hybrid printing, a 3D structure with multilayered circuitry can be achieved by printing the conductive and insulating structural material alternatively for each layer and connecting different conductive layers through a carefully designed inter-layer conductive path.

Alternatively, 3D circuits or conductive tracks can be achieved by using conformal printing via a multi-axis printing platform (typically 3 or more axes) (**Figure 9b**).^[159, 161] Conformal printing of MLMM electronics is a printing process where functional inks are deposited and patterned on a 3D freeform surface. Conformal printing can be done using various DW print heads such as inkjet printing and DIW print heads.^[159, 161] However, it is more commonly done using aerosol jet printing technique because of the high standoff distance between the nozzle and the substrate which makes it more tolerating to the non-uniformity of the substrate's surface. In comparison to

printing on flat surfaces, conformal printing is more complex as there are more design and fabrication considerations such as the accessibility of the print head to the surface of the 3D structures, toolpath planning, the alignment of the 3D parts to the print head, and the heating of the surfaces with arbitrary geometry for optimal printing.^[159] Although most of the work on conformal printing of electronics involves single material printing for conductive tracks only, however, the same knowledge that has been discussed in earlier sections can also be applied to achieve conformal printing of MLMM electronics.

On the other hand, 3D MLMM electronics can also be achieved using various 3D printing techniques using nanocomposite materials to form 3D self-supporting electronic structures (**Figure 9c**).^[160] It has been demonstrated in various works that free-standing electronic structures can be achieved using direct-ink writing, fused deposition modeling, stereolithography, digital projection lithography, powder bed fusion, and binder jet, just to name a few. Among these

Figure 9. (a) Fabrication of 3D MLMM components and multi-layered circuitry using hybrid printing technology [Reprinted with permission from ref^[127]. Copyright 2014 Elsevier], (b) Conformal printing of circuits on 3D curvilinear surfaces [Reprinted with permission from ref^[17, 162]. Copyright 2012 IEEE, 2011 Wiley], and (c) Fabrication of free-standing MLMM tactile sensor using DIW technology [Reprinted with permission from ref^[15]. Copyright 2017 Wiley].

techniques, DIW is found to be the more suitable for 3D MLMM electronics as the implementation can be done easily with 3D printers with multi-nozzles and multi-ink reservoirs. Instead of using solvent-based functional inks that normally have low viscosity, the fabrication of free-standing structural electronics using DIW techniques requires high viscosity functional inks as the extruded filament has to its shape to form the 3D structures.^[49] The nanocomposite functional inks are normally made of resin mixed with conductive filler such as silver nanoparticles, graphene, and carbon nanotubes.^[92, 116, 160] Despite the ability to form 3D electronic structures, the electrical property of the structure made from such nanocomposite inks normally is inferior to those solvent-based functional ink because of the existence of matrix materials such as the resin and binding agent.^[92] Nevertheless, it creates more design freedom in realizing sophisticated multi-material electronics architectures that can be useful in various applications such as sensors, batteries, and nanogenerators for wearables.^[49]

3. Pre-, Intermediate- and post-printing treatment for MLMM electronics

This section will cover various pre-, intermediate- and post-printing treatments that are commonly practiced for the fabrication of 3D-printed MLMM electronics. Various surface treatments and sintering treatments for MLMM electronics will be discussed. In particular, the compatibility of the sintering techniques (thermal, intense pulsed light, laser sintering and UV curing) for MLMM electronics will be discussed.

3.1 Pre-printing treatment

Preparation of the substrate surface is an important step for the fabrication of printed electronics. This is because the condition of the surface has a significant impact on the surface wettability, printability of the ink, the adhesion of the printed film, and hence the performance of the printed devices. The surface of the substrate is normally prepared by cleaning with an organic solvent such as ethanol or isopropyl alcohol (IPA) to remove the contaminants such as oil and dirt on the substrate's surface.^[163] Typically, this is done by immersing the entire substrate in an ultrasonic bath filled with an organic solvent for an extended period (typically a few minutes). During the cleaning process, the non-polar compounds such as oil and grease will dissolve in the organic solvent, whereas the particles such as dust on the substrate's surface will be removed by the cavitation bubbles induced by the high-frequency pressure wave produced by an ultrasonic

transducer (**Figure 10a**). Although adding surfactant such as soap to a water bath could also improve the cleaning performance, however, the substrate's surface may contain traces of residual surfactant left behind during the drying process. This may require an additional cleaning step with deionized water to remove residual surfactant from the substrate surface. On the contrary, the advantage of using an organic solvent such as ethanol or IPA is that there will not be any residual substance after the drying process and the wetted surface can be dried in a much shorter time due to the high volatility of the solvents. The drying process of the wetted surfaces of the cleaned substrate can be accelerated by either blowing with an air gun or placing the substrate in an oven.

As discussed in the earlier section, rough and pores-containing surface conditions (surface roughness in the range of micrometers) of the substrate can lead to strong capillary action and cause detrimental impacts on the printability of the ink and the performance of the printed components or devices. Therefore, substrates with poor surface condition should be treated first prior to printing. Treatments for reducing the surface roughness of a polymeric substrate include physical polishing, chemical polishing, and vapor polishing.^[129, 164] Among the treatments, physical polishing by grinding with sandpaper and chemical polishing by wet acid etching is the more popular surface treatments for smoothening the rough surfaces of polymeric substrates.^[129, 164] For surfaces that contain pores, the surface can be treated by coating an additional passivation layer as the pores can be covered by the passivation layer and result in a smoother surface that is suitable for printing can be produced.^[165, 166] For instance, a thick layer of polyimide ink can be coated on the irregular surface of a 3D printed part to create a smooth surface for subsequent printing of conductive traces.^[166]

Often for printed electronics, the low surface energy and high smoothness of the substrate cause ink printability and adhesion issues, respectively.^[129] Surface treatment and modification are normally done to improve the surface wettability, ink printability, and film adhesion to the substrate. Generally, surface modification techniques modify the surface in the submicron level to increase the surface area for bonding, increase surface energy, create reactive sites and chemical functional groups on the surface, and remove surface impurities. For polymeric substrate, the surface treatment and modification techniques can be categorized into physical treatment, liquid chemical treatment, reactive gas cleaning treatment.

For substrates with high smoothness, physical treatment can improve the adhesion of the film by increasing the surface area and generating more sites for mechanical interlocking (**Figure 10b**).^[129] The commonly used physical treatment include surface roughening with sandpaper and media blasting.^[129] Media blasting is a type of physical process that can eject pellets of solid materials onto a surface by a pressurized stream of gas. The effect of the surface roughening processes varies depending on the abrading material, the substrate material, the initial surface quality, and the process parameters. As the abrading process involves removal of materials from the surface, particulates made up of the removed substrate material and the abrading material can be found on the surface of the substrate. Thus, an additional cleaning step is required to remove the contaminants from the substrate's surface.

Liquid chemical treatment involves exposing the substrate's surface to a chemical in the liquid phase which can react with the substrate material to form new surface functional groups (**Figure 10c**). Chromic acid is often used for treating the polymeric surface as it can introduce oxidized functional groups such as hydroxyl, carbonyl, and carboxylic acid, and alter the surface morphology by increasing the surface area, thus increasing sites for mechanical interlocking.^[129] On the other hand, potassium hydroxide solution is found to be effective in improving the surface wettability and film adhesion of polyimide film.^[167] The treatment time and temperature have a significant effect on the degree of oxidation and etch depth. Compared to the reactive gas surface treatment method, liquid chemical treatment works well with substrates with pores as the chemical can penetrate deeper by the capillary action.^[129] However, liquid chemical surface treatments have several considerable downsides. For instance, the use of liquid corrosive chemicals for the treatment often leads to the generation of large amounts of hazardous waste, which leads to environmental issues. Besides, additional processing steps such as washing, rinsing, and drying will be required to remove the chemicals from the substrate's surfaces.

Reactive gas surface treatment is a type of dry surface modification technique that is used to remove organic contaminants from the substrate's surface either by the collision energy of gas molecules and decomposition of contaminants by chemical actions.^[129] The process involves the generation of reactive gases or species such as a plasma or radical molecules and the exposure of the substrate's surface to the reactive gases (**Figure 10d**). During the process, organic contaminants can be removed by decomposition when exposed to the reactive species, at the same

time, inducing surface roughness and creating new functional groups on the surface resulting in better film adhesion and surface wettability. Plasma cleaning and UVC treatment are the more established dry surface treatment that is widely used for printed electronics. Other existing dry surface modification methods include laser cleaning, X-ray cleaning, and ion milling cleaning, but they are less explored for printed electronics.

Plasma surface modification method requires the use of high-frequency voltages (typically in the range of kilohertz to megahertz) to excite the atoms in the gas at low pressure to create the plasma required for the surface treatment.^[129] Generally, a plasma system that can operate at higher operating pressure requires more power for a higher degree of ionization as higher voltage frequency (MHz range) is used, and it could result in the damage of substrate due to the microwave heating effect. As such, plasma surface modification is normally done at low pressure in order to use kHz range voltage frequency for reducing the power consumption and damage to the substrate. There are various variants of vacuum plasma surface modifications, namely low-pressure nitrogen or oxygen plasma treatment and atmospheric pressure plasma treatment. For low-pressure plasma treatment, the closed chamber for surface treatment is normally fed with either nitrogen gas or oxygen gas to create different reactive species catering to specific substrates. For instance, Li et al. have shown that N₂-plasma treatment can result in PDMS samples with more stable reactive sites compared to O₂-plasma treatment, which leads to a longer lifetime of the functionalized surface and posing lesser time constraint in a manufacturing process.^[168] In contrast, Yang et al. demonstrate that O₂-plasma treatment is more superior to N₂-plasma treatment when it comes to treating polyimide samples as they found that the adhesion strength for O₂ plasma-treated samples is higher than the ones treated with N₂-plasma.^[169] Although low-pressure plasma treatment is very effective in treating the surface of the substrate, however, its use in a roll-to-roll production line can be challenging as the process has to be done at low pressure with a closed chamber. As such, atmospheric plasma treatment has been developed to realize plasma surface treatment without the need for a closed chamber. Part et al. have demonstrated the addition of sulphur hexafluorine to the gas mixture containing nitrogen gas, helium gas, and oxygen gas for the atmospheric plasma surface treatment can lead to better metal adhesion to a polyimide surface due to an increase in the formation of C=O bond on the substrate's surface.^[170] It should be emphasized that the radical sites or the freshly generated functional groups can be quenched at a considerable rate by the components in the ambient air, leading to the loss in hydrophilicity of the substrate's surface.

Typically, the surface wettability of a treated surface will return to its original state within a few hours. It has been shown in certain cases, interestingly, the long-term hydrophobic recovery of a surface due to the change in surface configuration can be partially reversed by immersion in water.^[171]

UV irradiation surface treatment differs from plasma surface treatments in terms of the formation of different reactive oxygen species (ROS) (**Figure 10e**). Unlike plasma treatments which the main ROS is typically atomic oxygen, UV irradiation treatment normally generates singlet oxygen molecules as the main ROS, thus resulting in different functional groups compared to the plasma treatments^[172, 173]. It has been found that UV treatment tends to form C=O bonds, whereas plasma treatment can produce a wider variety of bonds, including C-O bonds and C=O bonds.^[174] The effect of the UV irradiation treatment on surface wettability and thin-film adhesion depends on various factors such as the intensity, wavelength, and exposure duration.^[175] For instance, Rye et al. have investigated the effect of UV wavelength (UVA, UVB, and UVC) on the adhesion strength of printed copper films and found out that only UVC light (wavelength: 254 nm) is able to improve the surface wettability and copper film adhesion on polyimide film.^[176] The improvement in film adhesion can be attributed to the formation of C=O bonds as well as the surface roughening as a result of the UV treatment.^[176] However, long UV irradiation duration (e.g., > 10 hours) should be avoided to prevent over-roughening and unfavorable changes in the mechanical property of the substrates^[175]. Interestingly, the UV irradiation treatment can be done with various irradiation sources, such as UVC lamp, intense pulsed light lamp, and a laser source.^[175-177] Unlike other surface treatment methods, selective surface treatment can be made possible with a UV laser source to tailor different surface wettability on a substrate.^[177] A major advantage of UV treatment is that it can be done in the atmospheric condition without the need for oxygen gas supply. **Table 5** shows a summary of different surface treatments and their advantages and disadvantages.

Table 5. Advantages and disadvantages of different surface treatments

Surface treatments	Objectives	Advantages	Disadvantages
Ultrasonic cleaning	Remove contaminants	• Simple process	• Does not improve surface wettability
Surface roughening	Improve adhesion	• Simple process	• Requires experience to achieve homogenous roughening • Additional cleaning steps are required

Liquid chemical immersion	Improve wettability and adhesion	<ul style="list-style-type: none"> Works well with substrates with pores 	<ul style="list-style-type: none"> Requires additional steps such as washing and rinsing Can damage the substrate
Oxygen plasma treatment	<ul style="list-style-type: none"> Remove organic contaminant Improve wettability and adhesion 	<ul style="list-style-type: none"> Dry process Applicable to almost all materials 	<ul style="list-style-type: none"> Not applicable to localized treatment Short lifespan of the modified surface
UV irradiation	Improve wettability and adhesion	<ul style="list-style-type: none"> Dry process Localized treatment is possible 	<ul style="list-style-type: none"> Short lifespan of the modified surface

Figure 10. Various pre-printing treatments. (a) Ultrasonic cleaning of substrate, (b) surface roughening using abrasive materials to create mechanical interlocking sites, (c-e) Various surface

functionalization techniques to improve surface adhesion and wettability, (c) liquid chemical treatment, (d) plasma treatment, (e) UVC irradiation.

3.2 Intermediate and post-processing treatments

There are various types of inks that are currently available for printed electronics, such as metallic nanoparticle inks, metallic organic decomposition inks, conductive polymer ink, dielectric ink, semiconductor inks, etc. Most of the inks used for printed electronics require further post-processing treatment to enhance the functionality of the printed film in terms of its electrical performance and mechanical integrity. For MLMM printing, as the process involves the use of different inks at different layers, it will require some intermediate processing steps to sinter and cure the materials. Often, the intermediate treatments are similar to those of the post-processing treatments. For convenience, the term “post-processing” will be used to represent both intermediate treatments and post-processing treatments.

The post-processing treatment is dependent on the type of ink used. For instance, nanoparticle inks require a sintering process to remove the organic stabilizer on its particle surface and to allow densification of nanoparticle film to occur. Organic polymer ink, on the other hand, requires a curing process to induce photopolymerization to allow cross-linking of the polymers. Some of the particle-loaded inks like graphene and carbon nanotubes inks are readily conductive as-printed, a heat treatment process at elevated temperature is normally used to remove the surfactant from the materials for enhancing the film performance. Similarly, metallic organic decomposition (MOD) inks, a solution-based ink that contains metal precursors dissolved in a suitable solvent, also require a sintering step to chemically reduce to its elemental state.^[178] The sintering or curing of the printed film can be achieved by applying either using thermal energy or electromagnetic wave energy to the targeted film materials. In this section, various sintering and curing methods such as heat sintering, laser sintering, intense pulsed light sintering, and UV curing that are commonly used for printed electronics and their applicability to MLMM electronics will be discussed. Other sintering techniques such as chemical sintering, electrical sintering, induction sintering, and plasma sintering will be omitted from the discussion as they are less commonly used and/or do not cause any detrimental changes to the film's morphology that can affect their applicability to MLMM electronics. For sintering metallic film, it is important to note that a pre-sintering step is required for electrical and induction sintering techniques to make the printed film slightly

Thermal sintering is a process whereby heat is applied to the samples by means of radiation or convection. Thermal sintering is normally done using a convection oven. During the sintering process, the samples receive heat from the surrounding hot air, and the entire sample including the substrate and all the printed films will eventually reach thermal equilibrium. As the sintering process normally involves the evaporation of solvent and decomposition of the organic compound, film rupture can occur at places where the emitted gases are trapped within the film.^[181] This can result in undesirable surface morphology such as cracks which can affect the electrical performance of the device. Gradually increasing the temperature to the desired sintering temperature can prevent rapid expansion of the emitted gas and allow sufficient time for the trapped gas to be vented. The residual stress on the printed metallic film formed during the cooling process may induce cracks due to the mismatch between the expansion coefficients of the metal film and the substrates.^[182] An advantage of the thermal sintering method is that the heating is generally homogeneous throughout the samples, which can yield more consistent devices. However, since the temperature is uniform throughout the samples, it is important to ensure that all the printed materials have similar thermal stability to avoid degradation of materials.

Alternatively, the sintering of printed films can be done by using photonic treatment. Photonic treatment involves the exposure of samples to the light energy and the absorption of the light energy by the printed film, which then leads to the heating up of the material. The subsequent sintering mechanism of a photonic treatment is similar to that of thermal sintering. For photonic sintering, laser sintering and intense pulsed light sintering are the two methods that are frequently used for printed electronics. The effect of photonic sintering of printed film generally depends on several factors, namely the fluence of light energy and the interaction of the light energy with the materials including both films and substrates.

Intense pulsed light is short-pulsed white flash-light covering the entire visible light spectrum and a small portion of ultra-violet (380 nm) and near-infrared light (950 nm).^[140] The white light is normally generated using a xenon lamp as xenon gas is the most efficient gas to convert electrical energy to light energy. As most metal nanoparticles exhibit surface plasmon resonance in the visible light region of electromagnetic waves, the metallic film will absorb the pulsed light energy

and result in localized heating. For example, silver and copper nanoparticles, which are used in most metal-nanoparticle inks, have maximum absorption at wavelengths of 420 nm and 590 nm respectively. Interestingly, most polymer substrates only interact with the UV region of the pulsed light and the duration of pulsed light irradiation is too short to increase the temperature of polymer substrates. This gives IPL an advantage to sinter metallic nanoparticle film without damaging the polymer substrates. IPL technique has been shown to be useful for sintering various kinds of inks such as silver nanoparticles, copper nanoparticles, and silver nanowires. However, it should be pointed out that the effect of IPL depends on various factors such as particle size, shape, materials, etc. Compared to the thermal sintering method, the printed film can be sintered in a much shorter time (typically a few seconds or shorter) and it can easily be incorporated into roll-to-roll manufacturing. Despite the advantages of IPL techniques, the sintering treatment is prone to film rupture and crack delamination. The short burst of energy received by the nanoparticle from the IPL causes the solvent and organic compound to turn into gases in a very short time. The internal vaporization causes the gas to rush out of the printed film, which could lead to film swelling or rupturing.^[140] On the other hand, the MLMM electronics with metal and polymer layer structure can be problematic because the localized heating of the metal film can cause a large thermal expansion mismatch between the materials, leading to cracks due to the high residual stress across the layers.^[157] As such, proper sintering optimization is required to control the crack development and prevent film failure for the fabrication of MLMM electronics. Interestingly, Kwak et al. have demonstrated using the back irradiation method for sintering film on the transparent substrate can prevent vapor trapping and film rupturing.^[183]

Laser sintering method uses a laser source to create a tightly focused light beam with a coherent lightwave to sinter the printed film. Unlike IPL technique which uses a wide spectrum of light wave, laser sintering method normally uses a lightwave with a single wavelength. As different materials possess maximum absorption of light at different wavelengths, laser sintering techniques can be useful for MLMM electronics to selectively sinter the targeted film without damaging other film materials. Besides the localized film heating capability, the position and power of the laser sintering spot can be digitally controlled to achieve selective spatial sintering which cannot be done using IPL technique. Another advantage of laser sintering method is that it can be easily incorporated into a roll-to-roll manufacturing process for mass electronic fabrication.^[179] The laser fluence and wavelength are found to be the main process parameters affecting the performance of

the printed film. Paeng et al. have investigated the use of laser with three different wavelengths (near-ultraviolet 405nm, green 514.5 nm, and near-infrared 817 nm) for sintering silver nanoparticle film. They found out that laser with shorter wavelengths (ultraviolet and green light) have shallower optical penetration depth, thus it can only result in surface melting, whereas infrared laser can fully penetrate a silver film with a thickness of 170 nm.^[139] In addition, a continuous laser is found to yield silver conductive trace with lower resistance compared to pulsed laser.^[184] However, pulsed laser offers better localized heating capability compared to the continuous laser due to the smaller heat affected zone.^[63, 185] In terms of MLMM printing, it should be noted that laser treatment can induce different surface morphologies hence different surface wettability, thus possibly affecting the printability of ink for subsequent layers.^[155, 156]

UV treatment has been found useful for both sintering nanoparticle ink for conductive films and curing of photosensitive polymer for dielectric films.^[153, 186] The process involves an extended period of UV light exposure of the samples which can be easily incorporated into a roll-to-roll manufacturing process with a conveyer belt system. The mechanism of UV sintering of nanoparticles involves the heating up of both the nanoparticles and the polymeric substrate by the absorption of UV light.^[114] However, UV treatment of polymer substrates may suffer damage as the polymer can be heated up easily by UV light. In most cases, UV treatment is used for dielectric curing for MLMM electronics printing. The optimization of UV processing parameters is very important as it can affect the surface roughness of the dielectric layer, which then leads to different surface wettability for the subsequent print layer.^[153] It has been found that the surface roughness can often lead to undesired defects of the printed components.^[134] For the case of multi-material inkjet printing system, in-situ curing of the UV dielectric material is found to yield higher roughness compared to post-printing UV treatment as the ink drops do not have sufficient time to merge to form a smooth layer.^[153] **Table 6** summarizes the advantages and disadvantages of different processing treatments.

Table 6. Advantages and disadvantages of different post-processing treatments

Post-processing treatments	Energy source	Advantages	Disadvantages
Thermal sintering	Hot air	<ul style="list-style-type: none"> Uniform heating throughout the sample 	<ul style="list-style-type: none"> Localized sintering is not possible Slow

IPL sintering	Xenon lamp (mostly visible light)	<ul style="list-style-type: none"> • Targeted sintering of printed film • Fast process 	<ul style="list-style-type: none"> • Can result in cracks without proper optimization
Laser sintering	Laser source	<ul style="list-style-type: none"> • Selective sintering is possible • Able to control the wavelength 	<ul style="list-style-type: none"> • Slower than IPL
UV curing	UV lamp	<ul style="list-style-type: none"> • Can be easily incorporated into the R2R manufacturing process 	<ul style="list-style-type: none"> • Can induce surface roughness

4. Potential and Challenges of 3D printed MLMM electronics

This section will discuss the existing 3D printed MLMM electronics, focusing on their applications and performances. In addition, the challenges for 3D printing to be widely adopted for MLMM electronics fabrication will also be discussed.

4.1 Potential applications of 3D printed MLMM electronics

In terms of 3D printed MLMM electronics, there have been many successful attempts to fabricate various electronics components and devices with different MLMM architectures to demonstrate the potential of 3D printing of MLMM electronics. For instance, passive components, multi-layered circuitry, active components, light-emitting devices, and energy devices. The following sections will outline the materials used, the fabrication methods, and the performance of the respective printed MLMM electronics.

4.1.1 Passive components

Various passive electronic components such as resistors, capacitors, and inductors have been attempted in the past, but they are mainly printed in a single-material coplanar format such as the interdigitated electrodes ^[187] and antennas ^[188]. The single-layer electronic design architecture limits the potential of the printed components, and thus not suitable for high-performance electronics. With the advancement in 3D printing of MLMM electronics, passive components such as capacitors and inductors can be printed in a multi-layered format, and thus, lifting the design constraint and unleashing the potential of printed MLMM electronics for high-performance electronics. The printing of passive components such as resistors directly on an existing circuit is a great example that showcases the potential of MLMM electronics to eliminate the use of interconnect and soldering step which are often required in conventional circuit boards ^[147].

For printed capacitors, various research groups have successfully attempted fully printed parallel plate capacitors using MLMM architecture^[147, 189-191]. The fabrication of the multi-layered parallel plate capacitors is mostly done using droplet-jetting based printing technique such as inkjet printing technique^[147, 189, 190] and other using filament-extrusion based technique^[191]. Correia et al. used the workflow as depicted in **figure 11a** to fabricate a parallel plate capacitor using an inkjet printer with silver nanoparticle ink for the conductive layer and poly (4-vinyl phenol). The MLMM printing techniques allow for the stacking of components on each other to increase the overall component performance. For instance, the capacitance of a stacked capacitors configuration exhibit 3.1 nF/cm^2 , which is higher than the capacitance of the single capacitor implementation (2 nF/cm^2).^[147] On the other hand, Flowers et al. demonstrated using the FDM printing technique for the fabrication of structural capacitors and the printed multi-layered parallel plate capacitors (**figure 11b**) achieved a capacitance as high as 150 pF .^[191] In addition, Hardin et al. have shown that it is possible to fabricate fully printed high-performance capacitors with a close-loop control scheme. The capacitors exhibit capacitance as large as 314 pF (at 1 kHz) and breakdown over 1000 V , which is much higher than what has been recorded in past literature for printed capacitors.^[190]

Inductors are conductors that are coiled to exhibit certain inductance. For printed inductors, they normally exist as spirally wound conductive tracks on a substrate that is flat. Often, this can lead to one end of the inductor situated inside the spiral loop and another one outside of the spiral loop. As such, connecting the printed inductors to a circuit often requires a jumper wire. With MLMM printing, dielectric layers can be printed atop of the spiral conductive track to form a layer of passivation layer for another layer of printed conductors (**figure 11d**).^[147] By doing this, the connection points of the inductors can be conveniently placed side by side with each other to keep the circuit neat. Correia et al. demonstrated fabricated a printed inductor with MLMM architecture that exhibits an inductance of about $5.5 \text{ }\mu\text{H}$ for an external size of $14 \times 14 \text{ mm}^2$.^[147] On the other hand, Flowers et al. showcased 3D printed inductor for wireless charging application that was fabricated using the FDM printing technique (**figure 11e**).^[191] Other than that, similar techniques can also be applied to fabricate passive components like printed antennas for radio frequency applications such as RFID tags.^[166, 192]

4.1.2 Multi-layered circuits

Multi-layered printed circuit board fabricated using conventional PCB methods allows for a more compact circuitry design. However, the fabrication of multi-layered circuits remains a challenge for additive manufacturing technology due to the limitations of the printing processes. Currently, there are only a few works that have successfully demonstrated the use of additive manufacturing techniques for the fabrication of fully printed multilayered circuits. For instance, Mikkonen et al. have used inkjet printing technology to fabricate a stretchable multilayered circuit using silver

Figure 11. (a-c) Fully printed capacitors and (d,e) inductors. (a) design of multi-layered parallel plate capacitor and the inkjet-printed capacitors ^[147], (b) parallel plate capacitor printed using filament-based extrusion technique ^[191], and (c) high voltage parallel plate capacitor printed using inkjet printing technique ^[190]. (d) Inkjet-printed multilayered inductor ^[147], and (e) 3D printed inductor for wireless charging with filament-based extrusion printing technique ^[191]. [Adapted with permission from ref^[147], ^[191] and ^[190] Copyright 2017, 2017, and 2019 Elsevier, respectively].

nanoparticle ink for the conducting layers and PDMS ink for the dielectric layers (**figure 12a**).^[193]

As PDMS is hydrophobic, they have used intermediate surface treatment, printing, and sintering treatment for each layer to ensure that all the inks can be deposited nicely onto the underlying layers. This developed printing methodology has great potential for any biocompatible or skin-conformable application, where selective additive patterning of silicones is desirable. On the other

hand, Saleh et al. have used all-UV curable inks approach to overcome the limitation of the mismatch in the thermal compatibility of each ink in the post-processing environment.^[186] Using UV curing technique, they are able to sinter both the silver nanoparticle conductive ink and tri(propylene glycol) diacrylated (TPGDA) dielectric ink. Using the developed printing technique, they have fabricated different kinds of multi-layered circuits within 3D structures as shown in **figure 12b**, which illustrate the design flexibility of the new generation MLMM electronic circuitry using 3D printing.

Figure 12. (a) Fabrication of stretchable multi-layer circuits using PDMS substrate, silver inks for the conducting layers, and PDMS ink as the dielectric layers [Reprinted with permission from ref ^[193] Copyright 2020 American Chemical Society]. (b) Various MLMM circuits using multi-material inkjet printing techniques [Reprinted with permission from ref ^[186] Copyright 2017 Wiley].

4.1.3 Active components

Active components such as transistors and diodes are some of the fundamental components for logic circuits that are heavily used in advanced electronics. Typically, these components normally have multilayered structures and require different materials such as conductors, semiconductors, and dielectric materials. Conventionally, the fabrication of high-performance active components is done using silicon wafer technology. However, with the recent advances in flexible and stretchable electronics, silicon wafer technology falls short compared to additive manufacturing methods. With AM technology, various active components have been shown successfully to be fully printed on various flexible substrates. Transistors,^[189, 194, 195] diodes,^[196] and microelectromechanical (MEM) relay^[197] are some of the examples of fully printed MLMM active components.

Transistor is an electronic component that consists of 3 terminals, namely the source, drain, and gate electrodes. Normally, both the source and drain terminals will be situated at the same layer with the gate terminal separated from them with an additional dielectric layer, and there are a few possible configurations such as top contact-bottom gate transistor, bottom contact-bottom gate transistor, and top contact-top gate transistor.^[50] **Figure 13a** shows a typical fabrication process for a bottom-contact, top-gate transistor.^[194] Chen et al. demonstrated the use of carbon nanotube ink for the semiconducting layer and ion gel ink as the gating material for the fully printed transistor.^[194] They have shown that the printed transistor exhibits good mobility ($10\text{--}30\text{ cm}^2/(\text{V s})$), good on/off ratio ($10^4\text{--}10^7$), and good current carrying capacity. On the other hand, Kim et al. fabricated flexible transistors on polyethylene terephthalate (PET) substrate (**figure 13b**) and have shown that the printed transistors exhibit ON/OFF current ratios of 10^6 with $I_{\text{OFF}} < 10^{-10}\text{ A}$ (or leakage $< 10^{-6}\text{ A cm}^{-2}$), saturation hole mobilities of $\sim 1.3\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$, threshold voltages of -0.3 V , and subthreshold swings of 70 mV decade^{-1} , which is comparable to those printed on rigid substrate.^[198]

Interestingly, Park et al. have fabricated a fully printed MEM relay with a free-standing conductive structure using inkjet printing technology (**figure 13c**). The process involves the printing of a sacrificial support layer for the top source electrode. Once the top conductive layer is printed and sintered, the sacrificial layer is then chemically removed to produce the free-standing source electrodes, which is the main switch mechanism for the MEM relay.^[197] They have put in place proper methodology to ensure precise control of the actuation gap, and the resulting devices showed low on-state resistance, immeasurably low off-state leakage, good switching speed, and lower voltage operation than many other demonstrated printed electronic devices.

Figure 13. Various active components. (a) Typical fabrication process of a multi-layered thin film transistor [Reprinted with permission from ref^[194] Copyright 2011 American Chemical Society]. (b) Aerosol jet printed flexible transistors [Reprinted with permission from ref^[198] Copyright 2013 American Chemical Society], and (c) Microelectromechanical relays and the fabrication process using inkjet printing with a sacrificial layer [Reprinted with permission from ref^[197] Copyright 2013 American Chemical Society].

4.1.4 Light-emitting devices

Light-emitting devices are devices that give off light when a voltage is applied. Generally, the construction of light-emitting devices is made up of at least three or more layers comprising of

different materials. For the simplest three-layered architectures, they normally consist of a cathode, an emissive layer, and a transparent anode^[199] In contrast, a more complex light-emitting device such as a light-emitting diode (LED) can have up to 5 layers, with two additional layers namely the hole transport layer and interconnection layer.^[149] Interestingly, the light-emissive devices normally require a layer of transparent conductive layers as one of the electrodes because the light-emissive layer is normally sandwiched between the anode and cathode. In many cases, poly(ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS) is normally used as the transparent anode as it is one of the most stable and electrically conductive solution-processed ink available for transparent electronics.^[149, 199] Kong et al. made use of extrusion-based 3D printing techniques to fabricate a 5-layered LED with an emissive layer made up of quantum-dot material (**figure 14a**).^[149] They successfully fabricated a functioning LED on a contact lens, demonstrating the potential of using 3D printing technology to fabricate electronic components directly on 3D structures. The fabricated LED can emit light with a brightness of 100 cd/m² at 4.5 V, which is comparable to those fabricated on a flat substrate. The complexity in the design architecture of light-emitting devices and the ability of 3D printing to integrate distinct classes of materials

Figure 14. Various light-emitting devices, (a) 3D printed quantum-dot LED [Reprinted with permission from ref^[149]. Copyright 2014 American Chemical Society] , and (b) light-emitting device fabricated with hybrid printing technique [Reprinted with permission from ref^[199]. Copyright 2019 Wiley]

fully demonstrate the potential of current additive manufacturing technology for the fabrication of advanced electronics. On the other hand, Zimmermann et al. have fabricated a fully printed large-area flexible light-emitting device on biocompatible substrates using hybrid printing technology (**figure 14b**).^[199] They made use of the inkjet printing technique to accurately deposit the silver nanoparticle ink and PEDOT:PSS as the cathode and anode respectively, whereas the active layer is deposited using the blade coating technique. Such technique not only significantly speeds up the fabrication of large-area light-emitting devices, but also shows the scalability and ease of integration of 3D printing techniques with the conventional techniques for mass production of electronics.

4.1.5 Energy devices

Compared to other electronic components and devices, printed energy device is still a relatively new and emerging research field. Energy devices can be subcategorised into energy harvesting devices and energy storage devices. Energy harvesting devices are devices that can harvest energy from the environment and convert the energy into electrical energy that can be used for other electrical appliances. In contrast, energy storage devices are devices that can store electrical energy in the form of electric potential energy or chemical energy that can be used when needed.

Various types of energy harvesting devices have been attempted using printed electronics technology, the common ones are the solar cells^[200] and piezoelectric energy harvesting device.^[201] For solar cells, the energy harvesting process involves the absorption of light energy from the environment and converts the energy into electrical energy through the active layer. Like the light-emitting device, solar cells possess multi-layered architectures, the different layers are the back electrode, electron donor layer, photoelectric layer, electron acceptor layer, and the top contact electrode.^[202] Similarly, solar cells also require a transparent electrode to allow the light to pass through it and enter the active layer. As of now, high-performance silicon solar cells are made from the conventional silicon wafer technology aided with screen printing technology to fabricate the electrodes.^[203, 204] In the past, there have been various attempts to 3D print different material layers of the solar cells such as aerosol jet printing of solar cell's active layers (**figure 15a**)^[200] and the current collecting gride for the solar cells.^[205, 206] Recently, Luo et al. reported fully screen-printed perovskite solar cells that could exhibit a performance of 22.8 mA/cm², 0.911 V, 0.753, and 15.64% for the short circuit current, open-circuit voltage, fill factors, and power conversion efficiency, respectively.^[207]

For piezoelectric energy harvesting devices, the process involves the conversion of mechanical energy into electrical energy when the piezoelectric layer is deformed. A typical piezoelectric device consists of a piezoelectric layer sandwich between 2 electrodes. The low voltage generation of a piezoelectric component is an issue as it is not of much use to most electronic applications. The issue is more obvious with printed electronics as it is challenging to stack many layers of piezoelectric components together in series to multiply the generated voltage. To address this issue, Prakash et al. have carefully designed the printing methods that create an interlayer connection

between the electrodes of the printed thin-film piezoelectric components (**figure 15b**) to allow all the printed piezoelectric components to be connected in series, thus multiplying the voltage.^[201]

Compared to printed energy harvesting devices, the field of printed energy storage devices or printed batteries just started receiving more attention in recent years. Energy storage devices can be categorized into batteries that have high energy storage capability and micro-supercapacitors that can give high power output. A fully 3D printed freestanding lithium-ion battery has been reported by Fu et al (**figure 15c**).^[208] They have made use of DIW technique to pattern the in-house developed graphene oxide ink for the battery electrodes and a solid-state electrolyte ink. The solid-state electrolyte ink, polymer composite containing a mixture of poly(vinylidene fluoride)-co-hexafluoropropylene (PVDF-co-HFP) and Al₂O₃ nanoparticles, also serves as an electrically insulating separator. The 3D printed LFP cathode and LTO anode exhibit stable cycling performance with specific capacities of ~160 mAh/g and ~170 mA/g, respectively.

In contrast, there is more work related to the fabrication of fully fabricated micro supercapacitor.^[209-211] Micro-supercapacitors typically consist of a pair of interdigitated electrodes (IDE) with an electrolyte layer covering the IDEs. Interestingly, carbon-based nanomaterials such as graphene and carbon nanotubes have been widely explored as electrode materials due to their high surface area.^[211] Liu et al. demonstrated the fabrication of a flexible multilayered micro-supercapacitor that comprises a silver current collector, an electrode layer made up of silver/polypyrrole nanocomposite, and a gel electrolyte (PVA-H₃PO₄) layer using screen printing technique.^[212] The printed micro-supercapacitor exhibits an extremely high energy density (0.00433 mW h cm⁻²), remarkable flexibility (77.6% remained after 1000 bending times), and cyclic stability (82.6% remained after 10,000 cycles). The use of 3D printing techniques to fabricate micro-capacitors on flexible substrates such as polyimide and PET substrates fully demonstrates the potential of 3D printing and marks a breakthrough for the realization of fully printed wearable devices.^[211]

Figure 15. Various energy devices. (a) aerosol jet printing of solar cell's active layers [Reprinted with permission from ref ^[200]. Copyright 2011 American Chemical Society], (b) flexographic printing of serially connected piezoelectric energy harvesting components with cross-layer interconnection [Reprinted with permission from ref^[201]. Copyright 2014 Wiley], (c) 3D printing of a freestanding lithium-ion battery [Reprinted with permission from ref ^[208]. Copyright 2016 Wiley], and (d) screen-printing of flexible micro-supercapacitor [Reprinted with permission from ref^[212]. Copyright 2017 Wiley].

4.2 Emerging materials for printed electronics

Other than the commonly used functional inks for conductive traces and dielectric layers such as silver nanoparticle inks and polyimide inks, there are many other types of emerging inks that have been developed over the recent years catering for numerous applications such as transparent electronics, wearables, energy storage devices, and green and disposable electronics. These materials include 1 dimensional and 2-dimensional nanomaterials, quantum dots, nanocellulose-based composite, and biodegradable materials as summarized in **Table 7**.

Conductive 1-dimensional nanomaterials such as carbon nanotubes (CNT) and silver nanowires are very useful for transparent and flexible electronics due to their unique nanostructure. The conductive network that is made up of these slender 1D nanomaterials allows light to pass with the minimal obstruction which is beneficial for transparent electronics. Also, the strong and flexible nanostructure makes them suitable for wearables that require repeated flexing and folding.

2-dimensional nanomaterials are nanomaterials that have very thin layers (typically a few layers of atoms). They normally possess enhanced material performance in contrast to their build layered counterparts. For instance, graphene, which contains atomic thickness, tends to have high transparency and high electrical conductivity that makes it suitable for transparent and flexible electronics applications. MXenes, are 2D inorganic compounds that consist of transition metals such as carbides, nitrides, or carbonitrides. They have found applications in energy storage devices such as batteries and supercapacitors. Hexagonal boron nitride (h-BN), on the other hand, is a 2D dielectric nanomaterial that is useful for the fabrication of MLMM electronics that requires a dielectric layer such as transistors.

Quantum dots nanomaterials are semiconducting particles with a diameter in the range of 1-10 nm. They possess unique optical and electrical properties that are different from the larger particle due to quantum mechanics. In general, quantum dots inks have been extensively explored to be used for the fabrication of optoelectronics or displays. Typically, the light emission by the quantum dots is tuned by the particle size, with smaller particles emitting higher frequency electromagnetic waves. Currently, the key challenge for quantum dots inks is quantum efficiency. As of now, the commercially available quantum dots inks developed by UT Dots[®] can achieve a quantum efficiency of >50%.

Biodegradable electronics is an emerging field of printed electronics that focuses on the fabrication of green and disposable electronics with the use of biodegradable materials. In general, there are several approaches to develop biodegradable electronics with biodegradable inks, which are (1) using nanocellulose-based materials and (2) using polylactic acid, just to name a few. Readers are encouraged to read the review paper published by Hoeng et al. in which the use of nanocellulose-based inks for printed biodegradable electronics has been discussed comprehensively. Generally, nanocellulose-based inks are cellulose inks that are mixed with conductive fillers such as polypyrrole, poly(3,4-ethylenedioxythiophene), and silver particles. Due to the flexible nature of nanocellulose, it has found a broad range of applications in disposable flexible electronics. Similarly, polylactic acid, which is also a biodegradable material, has been reported to be used as dielectric materials and a binder for a tungsten-filled conductive paste. These works open a new possibility for the fabrication of disposable, low-cost organic electronic devices.

Table 7. Emerging printable inks for printed electronics

Inks	Materials	Properties	Main applications	Ref.
1D nanomaterials	• CNT nanocomposite	5 S cm^{-1}	Electrochemical sensor, electrodes, flexible electronics	[89, 213]
	• Silver nanowire	10^5 S m^{-1}	Flexible electronics and transparent electronics	
2D nanomaterials	• Graphene	$>500 \text{ S m}^{-1}$ and up to 40000 S m^{-1}	Transparent conductors, batteries' electrodes, sensors	[214-216]
	• Mxenes	$\sim 10,000 \text{ S cm}^{-1}$	Battery and supercapacitors	
	• h-BN	dielectric material	Dielectric layer for transistor	
Quantum dots	• CdSe* or CdS*	luminous efficacy: 165 lm/Watt quantum yield: $>50\%$	Optoelectronics and displays	[217-219]
Biodegradable electronic material	• Nanocellulose-based composite (CNC* + PPy*)	Up to 36 S cm^{-1}	Flexible electronics, Supercapacitor and biosensor	[220-222]
	• Polylactic acid-based composite	5000 S m^{-1}	Green and sustainable electronics, disposable electronics	

*CNC: cellulose nanocrystal, PPy: polypyrrole, CdSe: Cadmium selenide, CdS: Cadmium Sulfide

4.3 Challenges of 3D printed MLMM electronics

The fabrication of intricate multilayered device constructions proves highly challenging as it requires the considerations of various aspects including the materials, printing processes, post-processing treatments, and component design. First of all, there are plenty of inks comprising different material types such as conductors, semiconductors, and dielectric materials available for printed electronics, however, it is often difficult to identify the suitable solvent combinations for MLMM electronics.^[223] Also, as MLMM electronics involves different materials, it is challenging to ensure that all the materials available are not sensitive to the processing environment.^[223] Besides, as the rheological behavior of the inks could be different, printing them in the same layer could result in different layer thickness which could pose challenges for the subsequent layers.^[134] The non-uniformity of the obtained layers could result in faulty components and limit the maximum number of layers that can be printed per the curing condition of each material.^[147] Also, it can affect the yield of the production of MLMM electronics as non-uniformity can affect the consistency of the printed components. Short-circuit between conductors of top and bottom layers within an MLMM electronics due to the non-uniformity, cracks, pin-holes, and defects is another

challenge of 3D printed MLMM electronics.^[224] Although a thicker layer could be a potential solution, the electrical performance of the components, in turn, will become poorer.^[224] Kong et al. identified the key challenges for MLMM printing of high-performance electronics, namely the print resolution, the performance, and the yield of printed MLMM electronics.^[149] In terms of multi-layered electronics circuits, despite the advancement in 3D printed interconnects, there are still several challenges with ink plug vias that need to be addressed, namely (1) non-uniform conductivity across the interconnects, (2) outgassing of fully buried vias that were not fully cured, (3) shrinkage of ink when cured, and (4) the reliability of the interconnects upon thermal cycles.^[225] Other than that, the flexibility of using a variety of substrates for printed electronics also poses a challenge for electrical bonding such as soldering, adhesive bonding, and crimping because they have different requirements such as process temperature, the desired flexibility, the conductivity, and the adhesive strength.^[226] From the viewpoint of mass production of electronics, the throughput of MLMM electronics still pales in comparison to the conventional manufacturing techniques.^[211]

Conclusion

Additive manufacturing of electronics, or 3D printing of electronics, has been increasingly adopted for the fabrication of electronics. With the advancement in materials science, various materials can be made into printable inks that can be used for different 3D printing techniques. This has opened new opportunities for the creation of new electronic designs that can be manufactured using 3D printing. Despite the ability to 3D print these materials, various design and fabrication considerations need to be taken care of when printing these materials sequentially to form different functional layers. These considerations include the print resolution, the material compatibility, the mismatch in coefficient of thermal expansion of different materials, the surface wettability, the post-processing treatments and the resulting performance of the sintered film. Failure to consider these factors could lead to undesirable performance of the printed electronic devices. In brief, various fundamental electronic components and devices such as resistors, capacitors, inductors, antennas, transistors, diodes, relays, multi-layered circuitry, light-emitting devices, solar cells, and batteries have been successfully demonstrated using additive manufacturing technology. These breakthrough and successes in 3D printing of MLMM electronics have started to make people to rethink the ways of electronics fabrication. Not only that, 3D printing of MLMM electronics has received great attention in the fields of printed wearable electronics and multifunctional structures,

which the conventional electronic fabrication falls short of. However, various challenges remain for 3D printing of MLMM electronics such as identifying materials that are compatible with the MLMM electronics fabrication process, the consistency of the printed components, and the throughput of the 3D printing processes. All in all, more efforts still need to be done in order for 3D printing of MLMM electronics to be widely adopted by the electronic manufacturing industry.

Conflict of interest

The authors declare no conflict of interest.

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3D printing of multi-layered and multi-materials (MLMM) electronics has open new opportunities for unconventional electronics components and devices such as flexible electronics and wearable electronics. However, the fabrication of MLMM electronics is very challenging as various design and fabrication considerations need to be taken care of. This review outlines the key consideration for the fabrication of MLMM electronics and highlight the potential of 3D printing for future generation of electronics.

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3D Printing of Multi-Layered Multi-Materials Electronics: A Review

ToC figure

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