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A Guide to Printed Stretchable Conductors

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Tushar Sakorikar, Nikolas Mihaliak, Febby Krisnadi, Jinwoo Ma, Tae-il Kim, Minsik Kong, Omar Awartani, and Michael D. Dickey*

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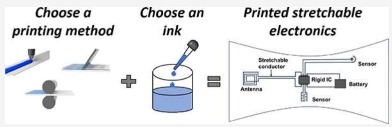
ABSTRACT: Printing of stretchable conductors enables the fabrication and rapid prototyping of stretchable electronic devices. For such applications, there are often specific process and material requirements such as print resolution, maximum strain, and electrical/ionic conductivity. This review highlights common printing methods and compatible inks that produce stretchable conductors. The review compares the capabilities, benefits, and limitations of each approach to help guide the selection of a suitable process and ink for an intended application. We also discuss methods to design and fabricate ink composites with the desired material properties (e.g., electrical conductance, viscosity, printability). This guide should help inform ongoing and future efforts to create soft, stretchable electronic devices for wearables, soft robots, e-skins, and sensors.



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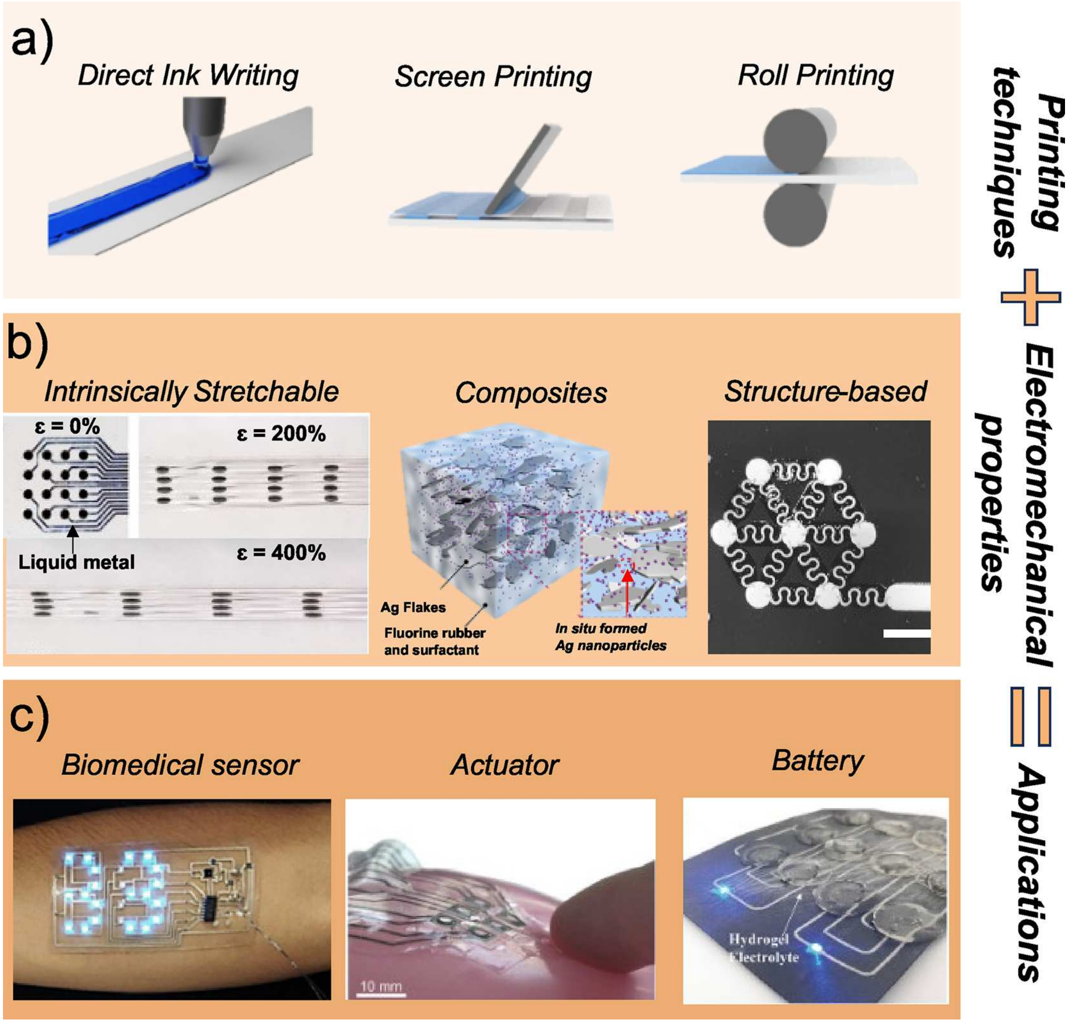

Figure 1. Printing techniques combined with special inks can produce stretchable conductors that lead to applications in sensors, actuators, and stretchable electronics. a) Schematic representation of commonly used printing methods. b) Strategies for stretchability: CNT electrodes (black circles) with intrinsically stretchable liquid metal interconnects. Reproduced with permission.[17] Copyright 2022, American Association for the Advancement of Science. Conductive composites composed of Ag flakes and fluorine rubber containing a surfactant. Ag nanoparticles were formed in situ between the flakes due to postprinting sintering. Reproduced with permission.[18] Copyright 2017, Springer Nature. Meandering shaped conductors are stretchable electrode due to their shape. Scale bar: 1 mm. Reproduced with permission.[19] Copyright 2018, Wiley-VCH. c) Applications of printed stretchable conductors: Biomedical sensor made from silver-liquid metal composite ink (Reproduced with permission.[20] Copyrights 2021, Springer Nature), actuator using carbon-based ink (Reproduced with permission.[15] Copyrights 2023, Wiley-VCH) and stretchable battery using electrodes composed of a stretchable liquid metal−silver-elastomer composite (Reproduced with permission[16] Copyrights 2022, Wiley-VCH).

1. INTRODUCTION

Stretchable electronic devices remain functional under deformation including elongation and bending. This distinguishes them from “flexible” devices that are merely capable of bending. Stretchability is important for emerging applications of electronics, such as devices that can conform comfortably to the body, sensors/electronic components for soft robotics, and electronics built into clothing or other unconventional substrates. There are many demonstrations in the literature such as stretchable displays,[1] implantable devices,[2] wearables,[3] and batteries.[4] Stretchable conductors are integral to these devices since they find use as interconnects (electrically conductive pathways), antennas, and electrodes.

Conventional methods to realize electrical conductors for electronic devices typically involve multistep cleanroom microfabrication processes like lithography, dry or wet chemical etching and vacuum deposition of materials.[5] Printed circuit boards (PCBs), can be fabricated inexpensively due to the high volume of PCBs. These days, even custom PCBs can be created inexpensively using these established tools. Traces are obtained lithographically in combination with chemical etching of copper sheets attached to laminates or substrates that are electrically insulating.[6] The overall process of manufacturing PCB has not been adapted for stretchable substrates nor is it a printing-based approach. Potential challenges of using existing PCB technology for stretchable devices include compatibility of stretchable substrates with the thermal and chemical processes uses for PCBs. Printing offers greater versatility and simpler process flows with lower overhead costs due to the simpler equipment. Prints can be achieved on large areas and on a variety of soft or unconventional substrates including textiles,[7] polymers,[8] and biological tissue[9] without the need for a cleanroom. Printing is typically additive; that is, material is only placed where it is needed without waste. Printing is also useful for rapid prototyping because of its ability to convert a computer drawing into circuits using laboratory (noncleanroom) based tools.

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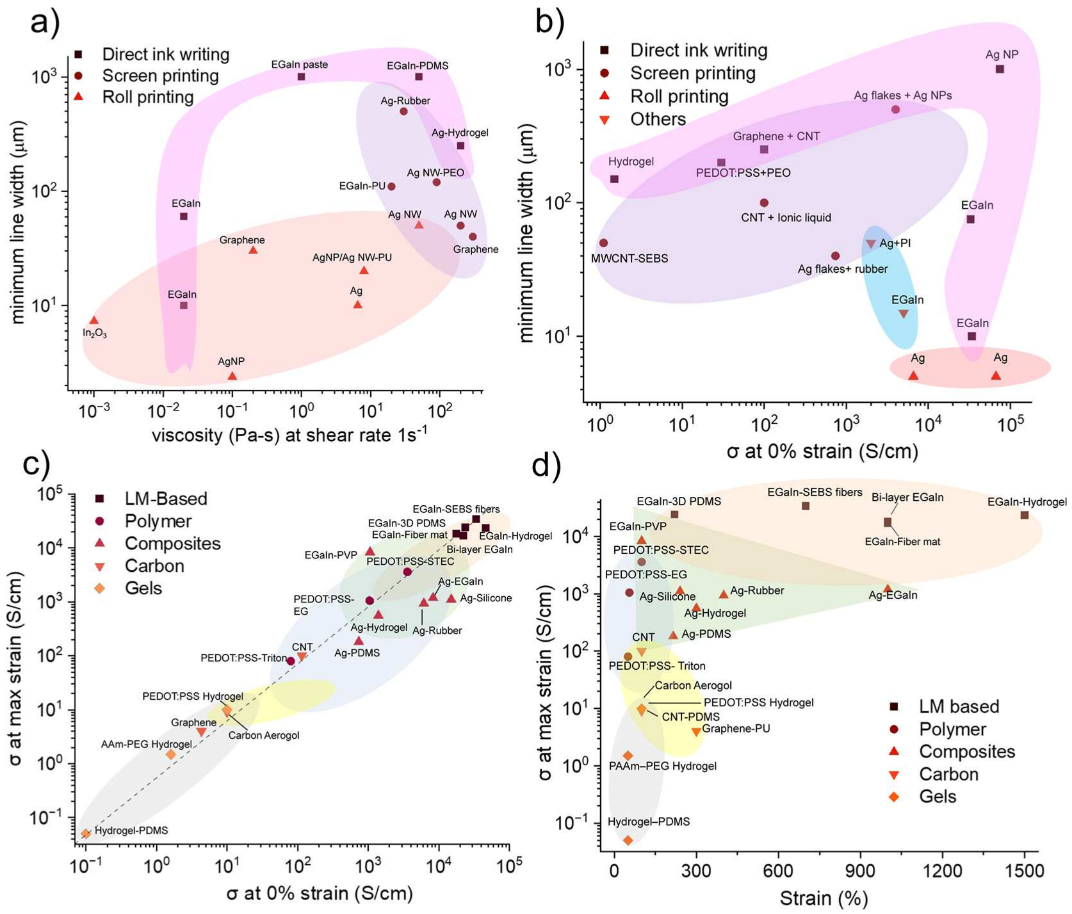

Figure 2. Ashby plots comparing various parameters of interest for printing. a) Minimum line width versus apparent viscosity for different inks categorized under different printing methods.[1] b) Minimum line width versus conductivity at 0% strain.[2] In (a−b), the shaded regions help guide the eye to cluster printing methods (pink for direct ink writing, purple for screen printing, red for roll printing and light blue for other printing methods). The shaded regions highlight demonstrated capability, but do not necessarily bound the physical possibilities for each method. c) Conductivity at maximum strain compared with conductivity at 0% strain and d) Maximum strain value for different materials inks categorized under five different material types. (The dashed line in c) represents strain insensitive conductivity of an ideal stretchable conductor.) In (c−d), the different categories of conductive materials are highlighted using shaded regions to guide the eye (orange for liquid metal (LM), green for composites, blue for polymers, yellow for carbon-based conductors and gray for gels). The references shown in a) and b) are listed in Table S1 and Table S2 and those in c) and d) are listed in Table S3 in the Supporting Information.

While the resolution of printing (1−100 μm) is worse than state-of-the art lithography (as low as tens of nm), the length scales are often sufficient for many applications such as interconnects between components, electrodes, antennas, and sensors. Furthermore, printing is limited in the sense that it cannot create the same quality (conductivity, density) of metal films deposited in more conventional ways, such as physical vapor deposition and sputtering. While printing can pattern a variety of conductors, we focus here on ones that are stretchable.

To print a stretchable conductor, the printing method and properties of the printed material should be considered. The properties of interest include printability, speed of print (area per time), resolution, registration/alignment of the printed material with other components on the substrate, cost, mechanical properties (stretchability), durability, electrical conductivity, and electromechanical properties (*i.e.* what happens to the resistance as the conductor gets strained). These considerations can be broadly categorized into two categories: printing method and electromechanical properties (electrical conductivity and stretchability). Examples of printing techniques include direct ink write printing, screen printing, and gravure printing (Figure 1a). The selection of the printing method is governed by factors such as resolution, speed, cost, and ease of use as well as the compatibility of a desired material with the printing method.

Stretchability may be an intrinsic property of the ink or achieved by clever structural or material design. Liquid metals, such as eutectic gallium indium (EGaIn)[10] are the only material we are aware of that can be deposited without any modification to form intrinsically conductive and stretchable structures (Figure 1c), although they need to be encased in elastomer to have elastic behavior. Conductive polymers[11] are generally rigid, but they can be blended with other polymers or additives to render them stretchable. Likewise, rigid electrically conductive materials can be rendered stretchable either by dispersing them as particles on elastomers or in elastomeric composites[12] or by patterning them in deterministic geometries, such as meandering traces[13] (Figure 1b). Several applications using stretchable conductors can be realized including biomedical sensors,[14] haptic interfaces[15] and energy storage components[16] (Figure 1c). Such applications are enabled by combining materials having desirable electromechanical properties (stretchability, conductivity) with an appropriate printing method based primarily on the rheological properties of the ink.

This guide is intended to assist researchers to identify and match the material behavior with a suitable printing technique. We propose a selection strategy and curate a collection of works that illustrate different printing techniques and stretchable conductors that are printable. We categorize the approaches and highlight their strengths and weaknesses. We focus on materials that can be printed and form stretchable conductors without the need to use the structural approach (i.e., deterministic geometry) shown in Figure 1b. Although we exclude the structural approach here, there are excellent reviews in the literature on this topic.[21,22] A major limitation of using deterministic geometry is that they require more area compared to the intrinsically stretchable materials, thus, limiting the device density. For more in-depth discussion of stretchable conductors including those that are not printable, the following reviews are good sources to consult.[23,24]

1.1. Definitions

As this guide discusses work that crosses disciplines, we define key terms that will be used frequently.

Printing. Printing is the controlled deposition of ink on a substrate into a desired geometry. The process can either be computer or manually controlled. The geometry of the printed structure is usually defined by depositing inks using computer aided dispensing (e.g., from a nozzle or an orifice), prefabricated masks, or features on a stamp.

Stretchable Conductor. Stretchable conductors are materials capable of conducting electrical current under mechanical deformation without loss of continuity. For any practical application, stretchable conductors should be elastic; that is, they should return to their original shape after strain. The range of conductivities vary for different classes of materials like metals (10^7−10^5 S/cm),[25] carbon-based composites (10^2−10^3 S/cm),[26,27] conductive polymers (10^2−10^3 S/cm)[28,29] and hydrogels (10^{-1}−10^2 S/cm).[30,31]

Resolution. Resolution is the minimum line width that is achievable for a given printing technique and ink. The parameters that influence/limit resolution depend on the technique. For example, droplet size for inkjet printing, mesh feature for screen printing, and nozzle diameter for extrusion printing.

Process of Selecting Printing Method and Material

In this guide we suggest the following steps to select a printing method and a stretchable, conductive ink for a particular application. We start by comparing and categorizing the various materials and methods for printing stretchable conductors to help provide a big picture comparison. We then more rigorously define and discuss these materials and methods in the sections that follow. There are several sequences by which a decision can be made of the best combination of method and ink; we suggest one here.

To select a printing method, one approach is to first define the desired resolution and conductivity. Figure 2a and Figure 2b summarize the resolution of representative printable stretchable conductors reported in the literature plotted against two different parameters. When considering print resolution, line spacing is another important parameter; the line-to-line spacing is often called "pitch". The ability to print conductors with reduced line spacing can lead to miniaturized stretchable electronic devices such as high density integrated circuits,[32] high frequency thin film transistors[33] and miniatured passive components (coils, resistors).[34] In Figure 2a it is plotted as a function of apparent viscosity of the ink at a shear rate of 1 s^{-1}. While there is not any apparent correlation between apparent viscosity and line width for the data set in Figure 2a, inks with higher viscosity values tend to a better print resolution.[35−37] Inks with higher viscosity spread less after printing, thus, improving the print resolution. Thus, Figure 2a should be interpreted as a resource to identify the range of typical viscosities used for each method, but not to show trends or correlations. This is important to help guide the selection of ink for a given task or to identify a target range during ink formulation. We pick a shear rate of 1 s^{-1} because it is a typical value experienced by an ink as it passes through the narrowest constriction in a nozzle or mask (e.g., assuming ≈1 mm/s average velocity through an orifice with a diameter of ≈1 mm). In general (and with some exceptions), gravure printing offers the finest resolution, followed by screen printing, and direct ink printing.

Each printing method has a range of viscosities over which the printing method works. In many cases, it is desirable to have a shear thinning ink so that it is easy to print (during which time the viscosity should be low while the shear rate is high) while also holding its shape after printing (during which time the viscosity should be high to help keep the ink from flowing until further steps can be taken to preserve the final shape, such as curing or encasing the printed structures). Stated differently, an ink may only be printable with a certain method depending on its rheological properties. For example, to screen print stretchable conductors, the apparent viscosity values for the ink are found to be >10 Pa-s as evident from Figure 2a. In contrast, inks with a broad range of viscosities (10^{-2}− 10^2 Pa-s at a shear rate of 1 s^{-1}) can be printed using roll printing and direct ink writing (Figure 2a). These range of viscosities can be used as a reference to modify inks. For example, a solvent like ethanol can be used to lower the viscosity of inks and then evaporate after printing. Similarly, additives like plasticizer can tune the ink viscosity for screen printing.

Many applications benefit from a combination of high-resolution and a high electrical conductivity. Among the printing methods, roll printing (e.g., reverse offset) offers the best of this combination for stretchable conductors (Figure 2b). However, there is always a trade-off: for example, reverse offset has stringent requirements on the rheology of material ink and the process of printing involves a three-step procedure using equipment that is not readily available in most academic laboratories. These steps will be discussed in section 2.3 under printing methods.

After selecting the printing method, a second step can involve selecting a material with desired electromechanical behavior that works with the chosen printing method. To choose from a palette of materials, electrical conductivities at both 0% and the maximum strain are plotted for commonly used printable conductive material inks in Figure 2c. The dashed line is a guideline for an ideal conductor behavior which is strain insensitive conductivity. Since some papers in the literature report only conductance, some of the values for electrical conductivities plotted here are back estimated using the sheet resistance/resistance value and the print geometry from the respective literature reports. It is evident, for instance, from Figure 2c, that liquid metal-based conductors provide the highest values of conductivities while staying in proximity to ideal conductor behavior. Another aspect to consider is the stretchability of the printed material. The user can select the material based on the desired value of strain and conductivity from Figure 2d. The liquid metal and its composites offer the combination of highest working range of strain and electrical

Table 1. Comparison of Loading Fractions and Electrical Conductivities for Different Composites

Filler Material	Optimal loading fraction	Conductivity (S/cm)	Comment	ref
CNT in LM matrix	CNT: 15 wt %	1.00×10^4	Increasing CNT content beyond 20 wt % lead to nozzle clogging	242
LM particles in polystyrene block copolymer	LM:74 wt %	2.00×10^4		209
LM particles in PDMS	LM: 90 wt %	1.00×10^3	At 90 wt % LM, the composite showed desired rheological properties of yield stress fluid and shear thinning	212
PEDOT:PSS in PEO	PEDOT:PSS: 50 wt %	~ 30	Increasing PEDOT:PSS > 50 wt % reduced stretchability, while decreasing below 50 wt % resulted in ~3 times increase in the sheet resistance	214
STEC in PEDOT:PSS	STEC: 45.5 wt %	2.50×10^3	Conductivity values saturated after STEC concentration >45.5 wt %	41
Ag flakes in fluorine rubber	Ag flakes:40 wt %	700	Increasing Ag flakes >40 wt % increased conductivity but reduced stretchability by more than 20 times.	243
Ag flakes in fluorine rubber	Ag flakes: 14 wt %	4.00×10^3	Ag flakes concentration of 14 wt % represents the value close to percolation threshold	244
Graphene in polyurethane	Graphene: 70 wt%	~ 22	Graphene concentration >70 wt % rendered the conductor brittle	40
CNT in PVDF	CNT: ~ 33 wt %	100		245

Table 2. List of Pros and Cons of Commonly Used Printing Methods for Stretchable Conductors

Printing Methods	Subtypes	Pros	Cons
Direct Ink writing	*Extrusion*	•Versatility: Can be used for a range of materials and viscosities •Simple setup flexible to changing geometries/patterns	•Sequential deposition: not great for large area unless multiple nozzles are used •Low print speed (1−10 mm/s)[246] •Print resolution limited by nozzle diameter and ink rheology
	Inkjet	•Great for dilute inks (1−20 mPa-s) without needing any binders[247] •Minimal material wastage[248] •Digital printing: geometries or patterns can be changed on-the-fly	•Low print throughput (10^{-3}−$0.5m^2/s$):[249] can only be increased with multiple orifices •Coffee ring effect during ink drying •Low print speed (10−50 mm/s)[250] •Orifice clogging •Print edges are not smoothWorks only on flat, smooth substrate
Screen printing	*Screen & Stencil*	•High throughput (2−3 m^2/s)[249] •Scalable •Economical and simple setup •Parallel deposition	•Versatility: New patterns require new stencils •Works only for inks with high viscosity. Binders or other additives are needed to make such viscous inks, that affect electrical conductivity •Works only on flat, smooth substrate
Roll printing	*Gravure printing*	•Very high throughput ($60m^2/s$)[249] •Scalable •Print edges are smooth •Parallel deposition	•Printing involves two steps: inking and deinking. This increases process complexity and failure modes •Inks are filled in cells or masters which needs to be changed for every design •Works only on flat, smooth substrate
	Reverse offset printing	•Scalable •High resolution •Parallel deposition	•The process involves three steps, adding complexity •Ink design has several constraints. For example, it should stay in a semidry state before transferring to the substrate •Works only on flat, smooth substrate
Others	*Aerosol jet printing*	•Works with wide range of viscosities (1 mPa-s to 1 Pa-s) •Maskless method •Can deposit materials on 3D structures	•Needs constant high pressure to generate aerosols •There exists a physical limit to nozzle diameter for a given droplet size, given by Stokes law and it scales as Q/D^3; where Q: flow rate (Q) and D: nozzle diameter[117] •Sequential deposition unless multiple nozzles are used
	Microcontact printing	•Low cost •Parallel deposition	•Each ink requires different modification of the stamp to ensure proper wettability •For finer features, stamp needs to made using standard microfabrication techniques •Deformation of the stamp •Few examples of stretchable conductors

conductivity under maximum deformation. The maximum deformation is governed by different factors for different types of inks. Liquid metal-based conductors for example are generally encased in an elastomer. The failure strain of the elastomer (e.g.,

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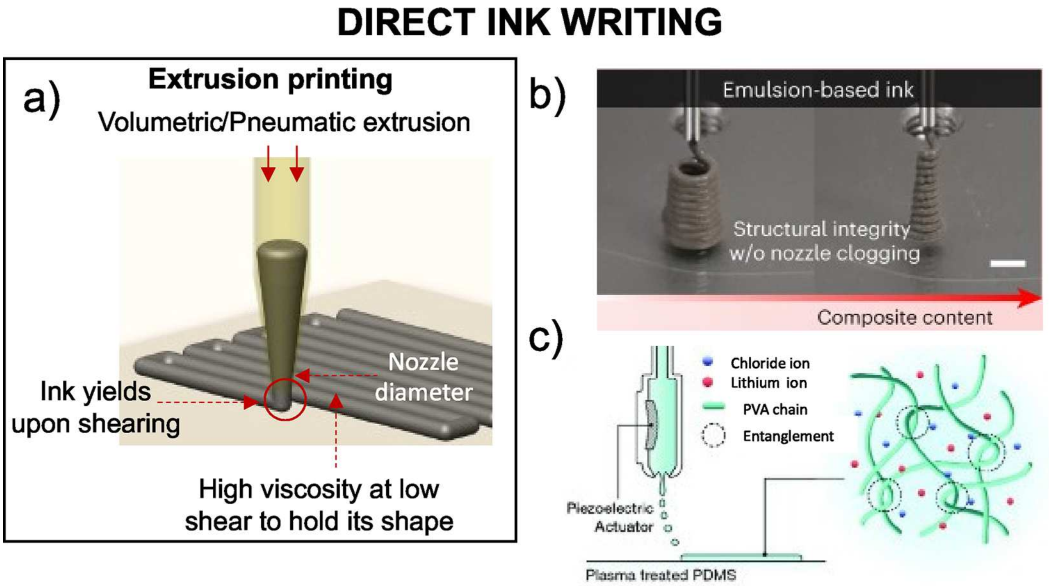

Figure 3. Direct ink writing involves dispensing an ink from a nozzle. a) Schematic illustration of important parameters considered for extrusion printing. b) As an example, emulsified Ag-CNT-PDMS composite ink with varying filler fractions. Scale bar: 1 mm. Reproduced with permission.[54] Copyright 2023, Springer Nature. c) Inkjet printing involves ejecting droplets from a nozzle. As an example, a hydrogel with ionic inks and PVA as a rheological modifier. Reproduced with permission.[55] Copyright 2020, Wiley-VCH.

800% strain for SEBS)[38] determines the maximum permissible strain for such conductors, although nonencapsulated liquid metal traces were shown to become unstable during strain when the diameter was ~10 μm.[39] For graphene-based conductors and conducting polymers, crack formation in the films is a common failure mode that limits their stretchability.[40,41] In the case of composites, the volume fraction of the conductive fillers and their spatial distribution affects the electrical conductivity under strain. Higher filler fraction (typically >40%, see Table 1 for more details) improves the conductivity but renders the composite brittle and more prone to cracking,[42] and therefore, a trade-off needs to be made. Reorganization of conductive fillers is another factor that affects the conductivity of composites under strain. When stretched, conductive fillers undergo a reorganization that changes the percolation path length which results in variation in the conductivity.[43]

Using this suggested approach, the final step involves iterating or compromising to resolve the (sometimes) conflicting requirements from step 1 and step 2, which may involve trade-offs. The second and third section of this guide provides a summary of each printing method and material ink type with their processing methods to make them printable and their limitations. The order presented here is by no means the only approach for a user to follow. The first step can also be selecting the desired electromechanical behavior and then identifying a suitable printing method.

Additional Considerations

In addition to printing methods and ink properties, additional considerations may be important. For example, from this analysis, liquid metal appears to be the most favorable stretchable conductor, but there are other factors to consider such as cost, adhesion to the surface, conductance vs conductivity (the former depends on geometry and the latter is an inherent material property), and print speed. In particular, the cost is an important consideration for manufacturing, but is rarely a consideration discussed in academic papers in which single prototypes are built; thus, we did not include it in our analysis despite its importance. For more information on the aspect of cost of printed electronics, readers are directed to some useful references.[44−46] While covering all of these additional considerations in depth is outside of the scope of this review (and often not discussed in academic papers), we do mention such additional considerations at various points in this review whenever possible.

2. PRINTING METHODS

Here, we focus on the most common methods of printing: direct ink writing (the term we use herein while noting it is sometimes called "direct-write" in the literature), screen printing, roll printing (gravure, reverse offset printing), aerosol jet printing (AJP) and microcontact printing (μCP). Of these methods, AJP is relatively new and μCP has not been used much for printing stretchable conductors; thus, these two patterning methods will only be discussed briefly under the heading "others". To help the reader assess and select a printing method, we have also included a table (Table 2) that lists pros and cons of printing methods discussed here.

2.1. Direct Ink Writing (DIW)

Direct ink writing (DIW) or "direct writing" creates patterns by extrusion of a filament onto a surface via a nozzle.[47,48] Though this is a generally agreeable definition, some works in the literature have also included inkjet printing under DIW which dispenses droplets rather than filaments.[49] In this review, inkjet printing will be categorized as a subsection under DIW to distinguish it from DIW methods that extrude filaments.

Extrusion Printing. Extrusion is the process of pushing material through a nozzle to produce filaments. The filaments can be patterned on a surface by moving the nozzle relative to a substrate. Most extrusion printer setups (such as in academic research laboratories) lack an integrated continuous feed system and therefore typically contain a single reservoir of material connected to the nozzle. There are several methods to push ink out from the nozzle. Volume-controlled dispensing can be done using syringe pumps in which movement of a plunger defines the precise amount of dispensed ink, while pressure-controlled dispensing can be achieved pneumatically. To integrate a printer system with a syringe pump, open-source firmware such as Marlin is useful.[50] Marlin controls the printer hardware (nozzle movement, bed leveling) and accessories like heaters and sensors. Furthermore, Marlin firmware is compatible with most of the commercially available 3D printing platforms used for extrusion printing. Other sources of pressure include thermal expansion[51] and Lorentz force in the case of conductive fluids.[52] Screw/auger extruders can help uniformly mix the ink, efficiently transfer thermal energy (important for materials that only flow at elevated temperatures), and to impart mechanical torque to push the ink from the nozzle.[53]

To ensure continuous extrusion of a filament, it is imperative to tune the rheological properties of the ink, such as viscosity, storage modulus, yield stress and surface tension.[56,57] In particular, inks with shear thinning or thixotropic properties are well suited for DIW because at high shear rates their viscosities decrease to allow flow through a nozzle. Sufficient storage modulus is necessary for good shape retention of the print once extruded, as shown in Figure 3a). As an illustrative example, it is possible to extrusion print PEDOT:PSS by modifying the rheology to impart shear thinning behavior. PEDOT:PSS shear thinning inks with an apparent viscosities in the range 100−200 Pa-s (at 1 s^{-1}) were found to be printable. Inks with viscosities less than ∼100 Pa-s showed poor shape retention upon extrusion, while those with viscosities greater than ∼200 Pa-s clogged the nozzle during extrusion.[58] Inks with a yield stress behavior can be printed. By applying a force above the yield stress, the ink can flow from the nozzle, but then once printed, the stress drops below the yield stress such that the ink holds the printed shape. This concept is utilized for printing liquid metals due to the yield stress of the oxide that forms on its surface.[59] Alternatively, inks can be designed to solidify after exiting the nozzle by using, for example, photocuring or solvent evaporation.

A key take-away is that the rheology of the ink is important for direct write printing. In addition to affecting the ability to extrude the ink and preserve the printed shape, one study showed that the rheology can also affect the resolution and roughness of the printed features.[60] Therefore, a systematic investigation should be done on any additive used to modify the ink rheology.[60] Toward this end, the range of viscosities as shown in Figure 2a can be used as a guideline for each printing method. Different strategies exist for modifying the ink viscosity. For example, increasing the solid content seems like a straightforward method to increase the storage modulus of the ink. However, it can also clog the nozzle. Adding an emulsifier can help stabilize the dispersion (i.e., avoid agglomeration of particles) to decrease nozzle clogging while retaining the required viscosity. This concept is illustrated in Figure 3b in which composite inks made from Ag-CNT-PDMS were emulsified using polyethylene glycol (PEG) and extruded with a print resolution of ∼86 μm. Due to the presence of PEG, even an increased solid content, as indicated by the label "composite content" in Figure 3b, does not lead to nozzle clogging.[54]

Liquids with low viscosity are easy to push out of a nozzle and form intimate contact with substrates, but there are several issues. First, they can continue to flow after being dispensed, which can either cause capillary breakup into droplets rather than forming a semicylindrical filament or cause spreading of the liquid (thus, worsening the resolution). Mechanisms to solidify the ink after it exits the nozzle can help alleviate some of these issues. For example, hydrogel ink can be extruded in a liquid state. After the ink exits the nozzle, photo or thermal curing can convert the ink into a solid.[61,62] In such cases, retaining the layer-by-layer structure of the printed material can be challenging and layers generally fuse, thereby, lowering the shape retention ability of the structures.[63−65] One approach to mitigate this issue is by adding additives like clay, nanocellulose and tannic acid to tune the rheology of the ink so it holds its shape after exiting the nozzle, as shown in Figure 3c.[66,67]

For stretchable conductors, the filament should also adhere to the substrate for reliable electromechanical behavior. In some cases, the ink may naturally adhere to the substrate. For example, the adhesion may be satisfactory if the ink contains elastomer of the same composition as the substrate, or prepolymer that can either react with the substrate or penetrate/entangle with a polymer substrate. Encasing the printed traces with additional elastomer that adheres to the substrate can also help avoid delamination issues. Alternatively, another strategy to achieve adhesion is using an additive that has inherently good adhesion to the substrate. Styrene block copolymers (SBCs) are one such class of materials that can double as an adhesive component in the ink, as well as a highly stretchable encapsulating matrix for the ink. Once printed, the SBCs in the composite ink also help adhere the printed structure to the substrate.[68]

Another consideration is how to control the movement of the nozzle. Ideally, the movement of the nozzle relative to the stage should be computer controlled. For example, G-code is a popular programming language for 3D printing. There can be some variation between the predefined pattern in the G-code and the actual printed pattern. This issue can occur when printing sharp corners due to the lag between the nozzle position and the filament landing point on the substrate. This lag - often referred to as "jet lag" - results in a deviation in the print geometry from a sharp corner to a curved corner.[65] One approach to address this challenge is to optimize the printing process parameters like print speed and gap height between the nozzle and the substrate.[69] If the surface has significant topography or is not globally flat, that adds an extra challenge to keep the nozzle at a controlled distance from the substrate surface. A common approach to deal with this challenge is to use a laser to measure the distance between the nozzle and the substrate.

Other than nozzle inner diameter (d_{in}), the print resolution (trace width, w) in DIW is significantly altered by the ratio of translational speed of the nozzle (v) and ink dispensing speed (u), and nozzle gap height from the substrate (h). Typical range of values of nozzle diameters for extrusion printing are 100−800 μm,[62,70,71] while nozzle speed (v) ranges from 0.4 to 50 mm/s.[70,72,73] These parameters need to be optimized to control the quality and geometry of the printed filament.[74,75] For example, increasing the ratio v/u offers a route to thin printed filament (i.e., increase the resolution) relative to the nozzle diameter, but this approach is ultimately limited due to capillary breakup of the filament. An additional complication is that viscoelastic materials exhibit mechanical hysteresis upon extrusion that results in the printed filament expanding.[76,77] The so-called "die swelling ratio" (d_{in}/diameter of the extrudate) captures this change in geometry, which have values >1. The effect of die swelling on minimum resolution can be overcome by adjusting the print velocity to help thin out the printed filament. A suitable range of velocities need to be identified specifically for each ink.[74,75] Higher print velocities could lead to breaking of filament if the velocity is higher than the "critical stretch

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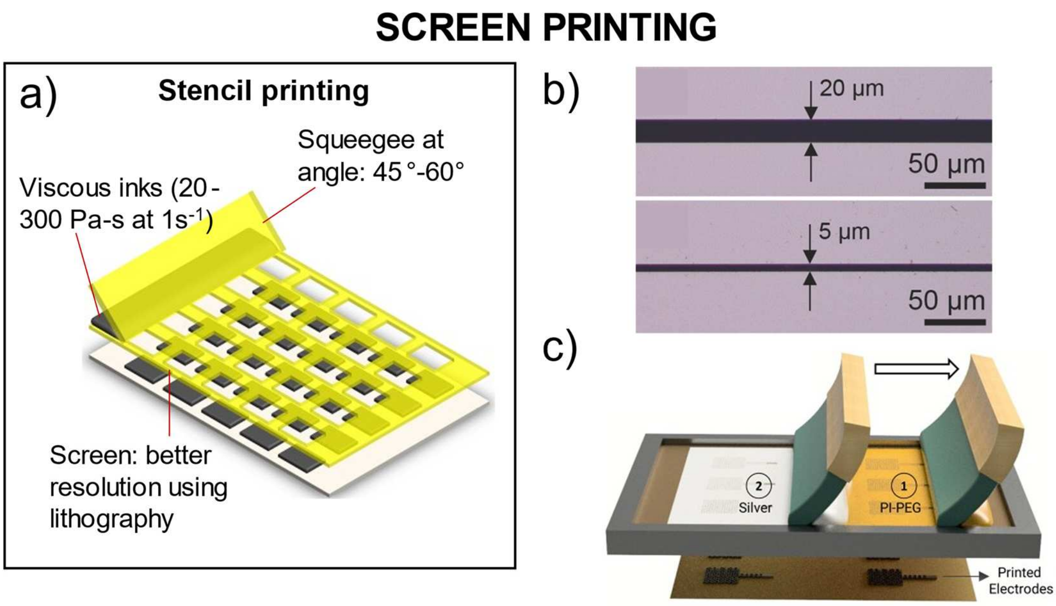

Figure 4. Screen printing typically involves squeezing ink through a predefined mask using an applicator. a) Process and components of stencil printing. b) Silicon stencils for high resolution screen printing. Reproduced with permission.[101] Copyright 2014, Wiley-VCH. c) Screen printing of Ag/AgCl inks for stretchable conductor and polyethylene glycol (PEG)−polyimide (PI) inks for the stretchable substrate. Reproduced with permission.[100] Copyright 2023, American Chemical Society.

rate".[78,79] Similarly, scaling down the nozzle diameters for better resolution has challenges that are dependent on the type of material inks. Nozzles with smaller diameters can lead to aggregation of conductive fillers in case of composites, and as a rule of thumb, the fillers should be ∼10× smaller than din to prevent clogging.[53] In case of LM, it is subjective to the method used for extrusion. When extruding LM pneumatically through smaller constrictions, maintaining sufficient pressure to prevent leakage of LM can be a big challenge.[80] For volumetric extrusion of LM, the limit on nozzle diameter is imposed by the ratio of gap height from the substrate (h) and d_{in}, which, if greater than 0.21, results in discontinuities in printed structures.[70]

Inkjet Printing. Inkjet prints ink by "jetting" droplets of the ink onto the substrate. Printing can be done either using a continuous stream ("jet") of droplets or drop-on-demand. Most of the commercially available inkjet material printers are drop-on-demand and the process is discussed briefly as follows.

When idle, the ink is held at the orifice (sometimes called "nozzle") of the printer by virtue of surface tension. When printing, droplets get pushed from the chamber. The most common method to eject droplets is to use an electric current pulse that actuates a piezoelectric membrane within the nozzle to generate a pressure pulse that dispenses droplets of ink from the orifice.[81,82] In bubble jet printing, the pulse of pressure is triggered by sudden heating that temporarily expands a bubble in the ink reservoir to push out the fluid from the orifice.[82] A challenge in drop-on-demand inkjet printing is clogging of the nozzle, which can happen as a result of ink drying at the tip, agglomeration of particles, or gelation of colloidal suspensions.[83,84] Material specific modifications are needed to solve the clogging issue.

Typically, inkjet printing works best for low viscosity liquids with low surface tension (not shown in Figure 2a). Although, it seems that a solvent with these two properties and a low boiling point should generate uniform print. However, such solvents lead to coffee ring effect, which results in deposition of material mostly at the periphery of the drop. A mixture of a low and a high boiling solvent used in an optimized ratio, has been found to address this issue. Due to the difference in the evaporation rates, a surface tension gradient is established causing a Marangoni flow that results in a reduced concentration gradient within the droplet as it dries.[85] A more detailed discussion on ink design can be found in this review.[82] Another consideration while designing ink especially for composites, is the size of the conductive particles such as AgNWs or CNTs. To prevent particle agglomeration that results in clogging, their size needs to be smaller than the nozzle, yet higher aspect ratio structures tend to be useful for stretchable conductors because they percolate better than low aspect ratio structures.[86]

As an example, consider liquid metal, which is difficult to inkjet print liquid metal because of its large tension and/or the presence of a native oxide on the surface of the metal. The oxide can be dissolved by having acid impregnated filter at the nozzle.[87] This approach requires specially designed orifice (700 μm diameter) and has not been implemented with commercial printers, which typically use smaller nozzles. Alternatively, liquid metal can be inkjet printed by suspending in a solvent some droplets with diameters smaller than the nozzle size. The resulting traces, however, are thin and not conductive due to the oxide that forms on the metal. Another approach to avoid clogging is to use particle-free inks. For example, silver salts can dissolve in printable solvents. After printing, the salts get reduced to a thin layer of metallic silver using an additional processing step.[88] The resulting traces have good electrical conductivity, but are thin and thus have poor conductance. They are also not stretchable.

In addition to avoiding clogging, the wetting dynamics of the ink on the substrate should be factored in during ink design by adjusting viscosity and surface tension.[82,89] The ratio of these two parameters are captured by the Ohnesorge (Oh) number.[90] Conventionally Oh is parametrized as Z ($Z = 1/\text{Oh}$), and numerical solutions reveal the condition for stable droplet formation: $10 > Z > 1$.[91] Utilizing this condition for droplet formation, salt-containing aqueous inks made from poly(vinyl

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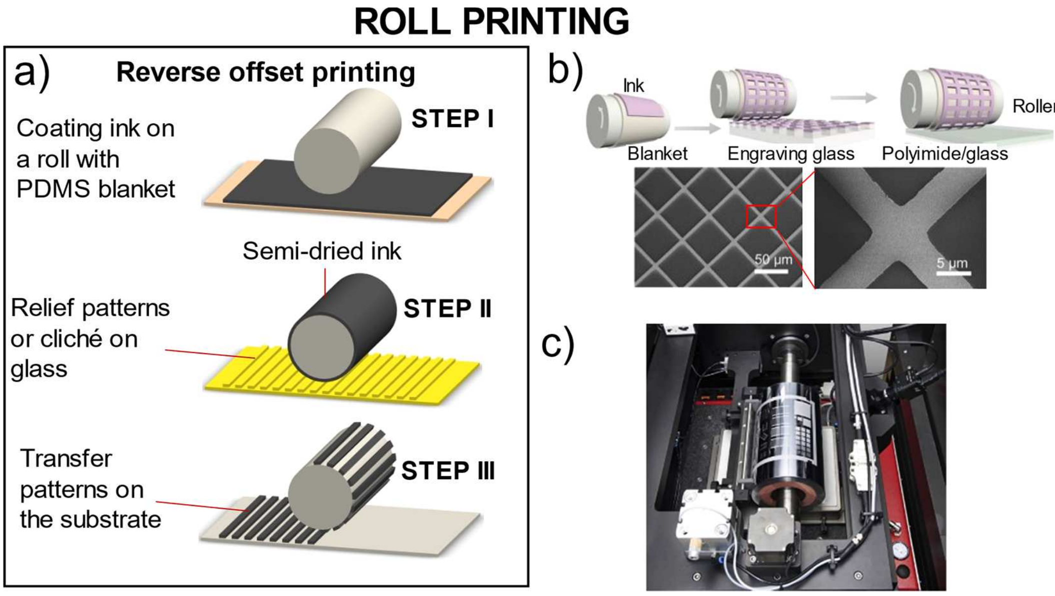

Figure 5. Reverse offset printing: a) Process steps involved to coat, pattern, and transfer ink. b) Reverse offset printing of stretchable Ag-containing mesh and SEM images of printed Ag mesh structure. Reproduced with permission.[113] Copyright 2018, Wiley-VCH. Gravure printing: c) Commercial material printer Challenger C600 used for gravure printing of CNT inks. Reproduced with permission.[114] Copyright 2018, Elsevier.

alcohol) (PVA) and a salt (lithium chloride) were formulated for inkjet printing (see Figure 3c). A droplet of volume ~40 pL resulted in a print resolution of 40 μm when inkjet printed on an elastomer, without the coffee ring effect–an effect that results from evaporation of solvent occurring faster at the edges of the droplet.[92] The printing resolution of inkjet printing is typically tens of microns, which is defined by the final spread of the drop (also known as footprint) when it is deposited on a surface. Inkjet printing also offers the ability of scaling up to improve the throughput by multinozzle printing. In this regard, multinozzle electrohydrodynamic inkjet printing, a variation of inkjet printing has become popular for high throughput printed electronics.[93,94]

2.2. Screen Printing

Screen printing involves pushing ink through a screen mask onto a surface. The screen mask consists of a screen mesh (a hatch pattern of nonwoven or steel wires) and an "emulsion" (polymer) or metal mask that defines the pattern of the ink by blocking it from passing through certain regions of the mesh. The screen is fixed on to a frame under tension and brought close to the substrate without touching it. Subsequently, the ink is spread across screen using a squeegee (or "applicator"). Only the openings in the screen allow the ink to flow through the mesh on the substrate. The mechanical tension of the screen and gap between the screen and substrate help the screen detach from the substrate after the squeegee passes over it, leaving only ink in contact with the substrate.[95,96] By tuning parameters such as viscosity and wetting of the ink, uniform prints can be obtained, while mesh dimensions (mesh opening, mesh wire inner diameter) and mask opening determine the resolution.[97,98] Generally, the mesh count ranges from 155 threads/inch to 400 threads/inch, translating to a printed line width of 150 to 50 μm.[97,99,100] The mesh serves two roles: (1) it helps to uniformly deposit the ink, and (2) helps support "island" structures in the mask (for example, concentric circles). Likewise, screen printing is reliable only for inks with high viscosity (see Figure 2a) as inks with low viscosity tend to spread laterally after passing through the screen. To prepare high viscosity inks, binders—typically polymers that can function as emulsifiers or adhesion promoters between different phases in in the inks—are needed which compromises the electrical conductivity of the ink.

Printing without a mesh is called stencil printing as shown in Figure 4a, although the terms "stencil" and "screen" printing are often used interchangeably.[3] Stencils are useful for several reasons: (1) they do not require a mesh, so stencils can be formed readily by laser or craft cutters, (2) not all inks can pass through a mesh, and (3) the openings in the stencil mask can be very small (for example, a stencil mask with an opening of 5 μm was patterned using photolithography as shown in Figure 4b).[103,104]

Squeegee pressure, speed, and angle are some additional "knobs" that can be used to improve the print quality of screen printing. Squeegee pressure and speed need to be adjusted based on the ink viscosity to ensure that sufficient hydrostatic pressure is maintained to transfer ink through the screen mesh.[105] Similarly, with the increasing squeegee angle (with respect to the screen) the hydrostatic pressure decreases and thus, reduces the amount of ink transferred on the substrate. Generally an acceptable range of squeegee angle is 45°–60°.[106] When screen printing straight lines for applications such as stretchable interconnects, orientation of features relative to the motion of squeegee should also be taken into consideration.[101,107]

Advantages of screen printing relative to other methods discussed here include high resolution (~100 μm is typical, but down to 5 μm is possible using special stencils), high throughput, low cost, and low material wastage.[97,108,109] Screen printing also offers flexibility to print conductive patterns and soft substrates. For example, polyimide (PI)-polyethylene glycol (PEG) composite as a soft substrate and Ag/AgCl-based conductive inks (minimum resolution: 150 μm) were sequentially screen printed using water as a solvent, as shown in Figure 4c. With a squeegee angle of 45° and a speed of 2 cm/s, PI–PEG substrates as thin as 1.2 μm were conformably coated on irregular surfaces.[100] Screen printing is used commercially to print flexible conductors and there is at least one stretchable,

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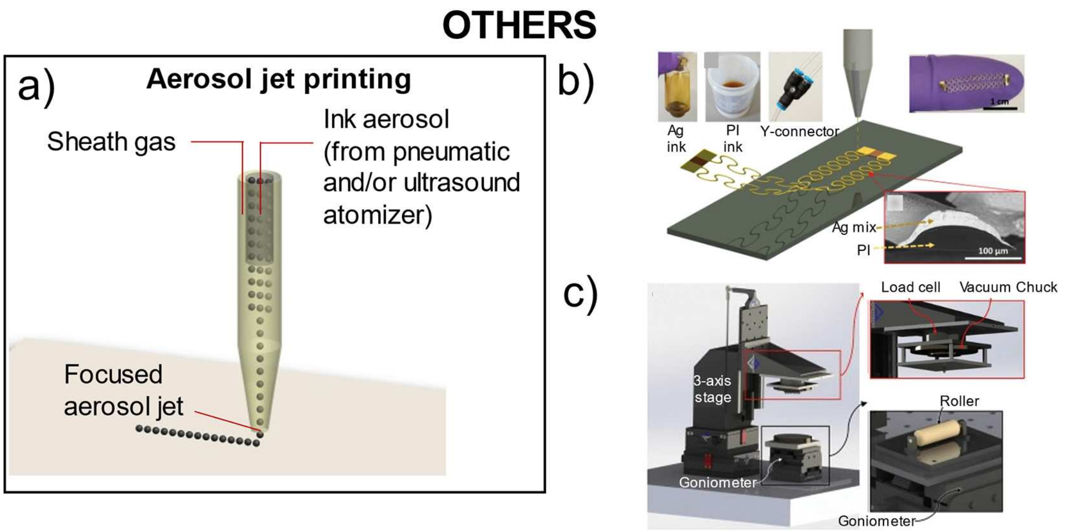

Figure 6. Other printing methods: a) Process illustration for aerosol jet printing, which uses a sheath gas to focus an aerosol of ink particles onto a surface. b) Ag and PI inks are separately atomized and then mixed to print free-standing stretchable conductors using aerosol jet printing (AJP). Here, the stretchability arises from the serpentine shape (deterministic geometry) since the polyimide itself is not stretchable. Reproduced with permission.[122] Copyright 2019, Wiley-VCH. c) Setup for microcontact printing of eutectic gallium indium (EGaIn) on an elastomer. Reproduced with permission.[123] Copyright 2019, Wiley-VCH.

screen-printable ink on the market (DuPont ME604). Figure 2c (battery) shows the types of stretchable conductors that have been reported in the literature using screen printing. Despite its advantages, screen printing also has some limitations. Other than the limitation offered by the range of permissible viscosity values as discussed before, the print resolution is limited by mesh opening (~40 μm).[101] Although, this can be overcome by lithographically defined silicon stencils over smaller areas, it limits the scalability of the process. Further, the process does not allow the change of patterns on the fly.

2.3. Roll Printing

Reverse offset and gravure printing typically use a rotating cylinder (roll) to transfer an ink onto a substrate brought into contact with the roll using another rotating cylinder (drum). Given these commonalities, these two methods are discussed in the same section, which we call "roll printing". Flexographic printing[110] is another type of roll-based printing, but among them, gravure and offset are generally used more frequently for printing stretchable conductors. A major advantage of these methods is high throughput printing with micrometer-scale resolution.[37,111] Also, roll printing can be used for printing nanomaterials like AgNWs with large aspect ratios (length >10 μm) without the issue of clogging as observed in nozzle-based printing methods.[112] Disadvantages include a larger upfront cost to obtain the patterned rolls, the inability to rapid prototype, nonidealities during the process of wiping by a doctor blade (for gravure)[37] and the narrow requirements for the properties of the ink (tension, rheology); the latter is true for reverse offset methods that use semidried films, whereas gravure methods work over a wider range of viscosities. Although not discussed in detail, it is useful to note here that flexographic offers a low-cost approach compared to gravure and reverse offset by utilizing relief features obtained by UV exposure of photopolymer films.[110]

Reverse Offset Printing. Reverse offset printing (ROP) consists of three steps. First, ink coats a rotating drum and dries to a semidry state. Second, the drum contacts a patterned substrate called a cliché. The purpose of this step is to remove the parts of the ink that are not required by utilizing adhesion of the semidried ink to the relief patterns on the cliché. As a result, patterns of ink remain on the drum. The semidry state is an important criteria as partial evaporation of the solvent changes the tackiness of the ink which helps in the selective removal using cliché and subsequent patterning on the substrate.[111] In the final step, the drum presses against a substrate to transfer the patterns. The process is illustrated in Figure 5a. A PDMS or a polymer liner (also referred to as "blanket" in the literature)[111] on the surface of the drum helps facilitate the transfer as schematically shown in Figure 5b[113] A key constraint of this process is the ink drying process. If the ink is not sufficiently dry, the pattern can smear. Conversely, a fully dried ink will not stick to the cliché in the second step. A careful selection of solvent and methods for drying is helpful in tackling this issue.[111,113,115]

Gravure Printing. The "image carrier" or "gravure roll" is a cylinder with an engraved metallic/ceramic surface. The engravings comprise individual cells that are filled with ink as the pattern rotates through an ink reservoir. A doctor blade scrapes off excess ink from the roll such that only ink in the cells remains. The gravure roll then rolls on the substrate, which is compressed in place between the gravure roll and an impression roll, allowing the ink pattern to transfer onto the substrate. Aside from the specifications of the gravure roll (e.g., cell depth and size, material), the quality of ink transfer depends on factors such as the applied pressure between the gravure and impression roll, rolling speed, relative wettability of ink on the gravure roll and substrate, and ink viscosity. Substrates are often treated with oxygen plasma to improve ink transfer.[37,116] Figure 5c shows the *Challenger C600*, a commercial roll-to-roll gravure printer which has a minimum engraver/cell depth of 8 μm. Carbon nanotube-based inks were printed using this system on a PDMS substrate with a print resolution of 20 μm.[114] Creating the engraved features on the surface of a cylindrical roll is nontrivial and thus, typically done professionally. The process of making rolls has evolved from mechanical to diamond stylus engraving, laser engraving, and electrochemical etching, and now photo-

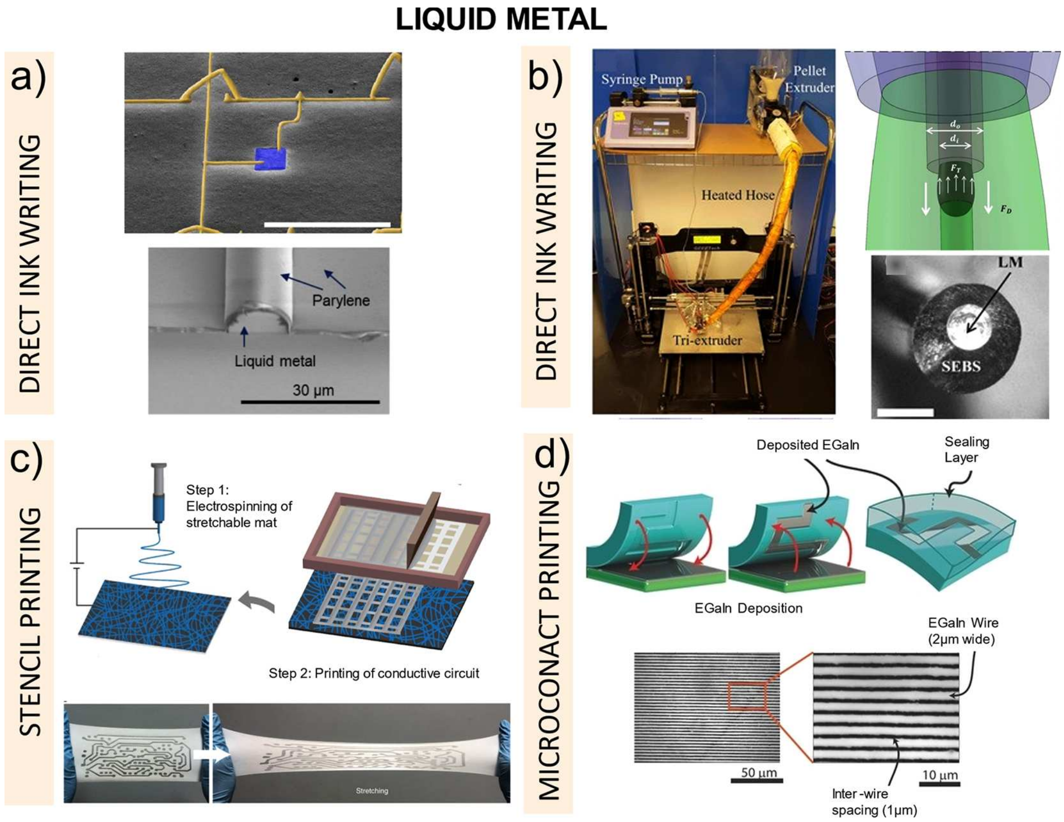

Figure 7. Strategies to print liquid metal. a) Pneumatically controlled DIW of liquid metal wires, shown in yellow false color (scale bar: 300 μm), encapsulated using a polymer deposited postprinting (scale bar: 30 μm). Reproduced with permission.[133] Copyright 2019, American Association for the Advancement of Science. b) Printing and simultaneous encapsulation of liquid metal can be done in single step by coextrusion with an elastomer (poly(styrene-*b*-ethylene-*co*-butylene-*b*-styrene) (SEBS)). Reproduced with permission.[134] Copyrights 2019, Wiley-VCH. c) Stencil printing of liquid metal on a sheet made from electrospun elastomer fibers. Reproduced with permission.[135] Copyright 2021, Springer Nature. d) Microcontact printing offers another route for maskless printing of liquid metal. Reprinted with permission.[136] Copyright 2013, American Chemical Society.

lithography is the mainstay to create finer features (<10 μm) that were not possible with the former two approaches.

2.4. Others

Aerosol Jet Printing. This method of printing was developed relatively recently compared to the others discussed so far. Although the process uses a jet of ink like inkjet printing, the process of creating the jet is conceptually different. Figure 6a shows the process of aerosol jet printing. The process begins by agitating the ink with ultrasonic waves to break it into smaller droplets (aerosols) in an atomizer. An inert carrier gas then transfers the aerosol from the atomizer to the deposition head. The deposition head consists of sheath gas flow that directs and focuses the aerosols toward the substrate.[117,118] A major advantage of aerosol jet printing (AJP) is that it can work with a wide range of ink viscosities as long as the ink can be aerosolized.[119] It is also capable of printing across 3D surfaces, whereas some of the other methods (screen printing, roll printing, direct-write) work best on flat surfaces. The AJP can also be used to print conductive composites by using a combination of pneumatic and ultrasonic atomizers and the resulting aerosols when combined using a Y-connector, can result in free-standing stretchable conductors as shown in Figure 6b.[120,121]

Microcontact Printing. Microcontact printing uses a stamp with an embossed surface covered with ink. Contacting the surface to a substrate transfers the ink.[124] A stamp is the most important part of this process as it governs the resolution and efficient transfer of patterned ink. PDMS stamps are sufficiently soft to make conformal contact to surfaces to ensure effective ink transfer. However, PDMS presents challenges. Its hydrophobicity makes it difficult for certain inks to wet the recessed features. Yet various surface treatment methods are availed to tune the surface energy such as oxygen plasma treatment and silane treatment.[125] It also has a resolution beyond limited to above the micron scale as the mold is soft and therefore deforms during printing.[126] The deformation also affects "registration"; that is, the alignment of features across a substrate. PDMS composites provide a facile route for mechanical modifications of the stamp to prevent sagging of the stamp during printing over large areas or finer features. Using materials with higher Youngs modulus like silicon is another approach to make stamps that do not deform while printing, but stiffer materials do not conform to the target substrate and are more prone to dust that can prevent intimate contact.

PDMS molds can be created easily by casting and curing liquid silicone against a master mold, formed using standard microfabrication techniques like lithography. Although, the microcontact printing is not demanding in terms of the setup involved, for finer features it warrants precise stamp loading on the substrate and accurate alignment in case of multiple layers.[123,126] The precise loading and movement of stamp can be achieved by a combination of a 3-axis motorized stage, a goniometer and the displacement sensors as shown in Figure 6c.[123]

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3. STRETCHABLE AND CONDUCTIVE MATERIALS

3.1. Liquid Metal

The term "liquid metal" is used here to describe the bulk form gallium and alloys of gallium that are liquid at or near room temperature. These alloys, unlike mercury and other metals that are liquid at/near room temperature, have low toxicity and low vapor pressure, which makes them generally safe to use.[127] Liquid metal is characterized by low bulk viscosity (0.002 Pa-s, 2× that of water)[128] and high surface tension (∼620 mN/m, ≈9× that of water).[129] Despite having a water-like bulk viscosity, liquid metal is a non-Newtonian fluid due to the presence of a thin oxide (∼3 nm) layer that forms spontaneously around the surface upon exposure to oxygen which helps retain non-spherical shapes.[130,131] Metallic electrical conductivity (3×10^4 S/cm)[24] and a reconfigurable shape makes liquid metal suitable for stretchable electronics and among the best options in terms of stretchability and conductivity (cf. Figure 2).[10] The oxide enables shape retention for printing.[59] A recent review highlights to pattern liquid metal;[132] we briefly discuss printing-based methods here.

Direct writing is one of the more explored methods to pattern liquid metal.[137] Since liquid metal has low bulk viscosity, dispensing it is typically dominated by interfacial forces. Once there is sufficient applied stress, the oxide at the nozzle tip yields and liquid metal exits the nozzle. This can lead to uncontrolled flow if the pressure is not balanced appropriately.[137] The metal naturally exits the nozzle as a spherical shape, similar to water. Yet, shear driven printing can address this issue to form wires and other nonspherical shapes stabilized by the oxide skin.[59] Shear printing works by applying just enough pressure to create a meniscus of metal that protrudes from the nozzle. Printing occurs by using the motion of the nozzle to shear the metal meniscus against the substrate or by using tension by pulling the nozzle away after adhering the meniscus to the substrate.[138] This approach requires good adhesion between the oxide and the substrate; the oxide on the metal adheres to many surfaces and adhesion can be improved by using an oxygen plasma treatment to the substrate.[138] While some printing methods discussed in this review result in thin traces due to evaporation of carrier solvent, liquid metal can be printed directly as a bulk material; in fact, it is difficult to print very thin liquid metal structures. Due to capillary effects, the thickness of printed liquid metal structures tend to be similar in length scale as the width. Thus, in addition to having excellent conductance (a material property), printed liquid metal features also have excellent conductance, a property that accounts for geometry.

Printed liquid metal patterns or self-supporting structures for stretchable electronics often require encapsulation to prevent mechanical damage (Figure 7a).[133] Simply stated, printed liquid metal structures smear readily if contacted without encapsulation. Such encapsulation can be cast over metal patterns postprinting, but can also be introduced using coextrusion of polymer along with the liquid metal, resulting in a single step process to print stretchable conductors (Figure 7b).[134] "Embedded extrusion printing" of liquid metal is another approach to direct write encapsulated liquid metal in a single step. Embedded printing is performed by extruding liquids in 3D space in a support bath that holds the shape of the print together. Yield stress fluids are preferred for the support bath because they help hold the shape of the printed metal while allowing the nozzle to pass through the bath. A combination of a yield stress fluid with sufficient critical yield stress and print speed ensures printing of bead-free continuous filaments of liquid metal.[139]

Stencil printing is another way to print liquid metal. In our experience (unpublished), screen printing is very difficult with bulk liquid metal because the oxide prevents the metal from flowing through the mesh; when pressed with a squeegee, the metal simply flows along the path of resistance, which is typically not through the mesh (this problem might be addressed by using a mesh that the liquid metal "wets"). However, there are several demonstrations of using stencil printing to pattern liquid metal. For example, printed liquid metal patterns were obtained on a porous electrospun SBS substrates and encapsulated by electrospinning another layer of SBS on top of the stencil printed metal (Figure 7c). This sequential process can then be repeated to obtain stacked layers of stretchable liquid metal conductors for multifunctional monolithic electronic devices (this work uses the word "stencil", but there are insufficient details provided to determine if the stencil is actually a "screen").[135] It is possible to stencil print liquid metal by either rubbing the metal back and forth over the stencil until it adheres to the substrate or by spraying the liquid metal across the stencil as an aerosol.[140]

Liquid metal can also be printed by a variation of microcontact printing which is a contact deposition method that generally uses stamps with relief features made from PDMS to print. There are at least two demonstrations of patterning liquid metal that fall within the spirit of microcontact printing.[123,136] In one case, droplets of liquid metal from a donor substrate are contacted by a PDMS slab and then transferred to a target substrate using a motorized stage to achieve a resolution of ∼340 μm. This approach transfers drops serially and does not use a topographic mold.[136] Thus, it deviates slightly from the conventional definition of microcontact printing. In another case, a topographic mold is pressed against a film of liquid metal such that the metal flows into the recesses of the topography (Figure 7d).[123] In this case, the metal stays in the mold and is not transferred to a target substrate. Thus, to date, there has not been a conventional demonstration of conventional microcontact printing in which a liquid is transferred from one surface to another using an elastomeric mold to facilitate the transfer. One of the challenges is ensuring adhesion of the metal to the PDMS stamp. This requires sufficient stress needs to be exerted on the liquid metal to break the surface oxide.[130] Thus, it is difficult to get the metal to flow into the relief features on a PDMS mold; when pressure is applied, the metal will simply flow along the path of least resistance, which unfortunately, is often not into the relief features. This issue can be minimized by using a very thin film of liquid metal for inking the molded stamp as shown in Figure 7d. Once printed, the features are then encapsulated with an elastomer (Figure 7d).[136] Features up to 2 μm line width can be obtained using this method as shown in Figure 7d.

There are many other interesting ways to pattern liquid metals that do not qualify as printing, but nevertheless have many of the desirable attributes of printing. For example, liquid metal can be injected into channels or selectively wetted to metallic traces. We limit our discussion to printing techniques here, but point the reader to other resources for ways to pattern liquid metals.[141,142] Finally, one open question is how to best make electrical contacts to liquid metals because of their ability to embrittle certain metals such as aluminum.[143] Recently, it was found that a composite of LM with Ag flakes in the presence of a polymer (styrene-isoprene-rubber), and optimized conditions

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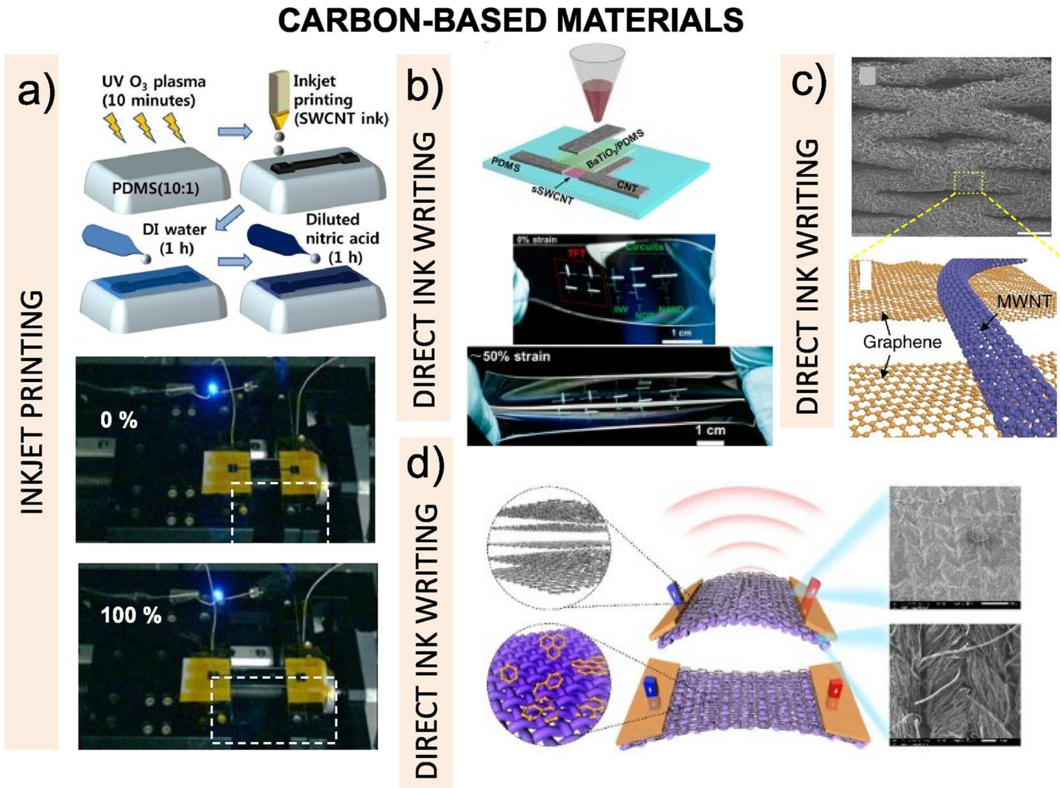

Figure 8. Approaches to print carbon-based stretchable conductors. a) Inkjet printing of CNT film on an elastomer followed by acid treatment to improve conductivity. Reprinted from[150] with the permission of AIP Publishing. b) Stretchable CNT interconnects and source drain electrodes in a thin film transistor printed on a PDMS substrate using direct ink writing. Reprinted with permission[151] Copyright 2016, American Chemical Society. c) Direct ink writing of graphene inks on a stretchable textile resulting in a conformal coating of textile fibers with multiple graphene layers. Reprinted with permission[152] Copyright 2023, American Chemical Society. d) Extrusion printed hierarchical structure with a unit element composed of graphene sheets with CNT as a rebar-like reinforcement. Reproduced with permission.[153] Copyright 2017, American Chemical Society.

of mixing, results in a stable electrical contact with desired electromechanical properties.[144] Alternatively, tuning the PEDOT to PSS ratio to improve the wettability of LM on the PEDOT:PSS surface, is another strategy for obtaining stable LM based stable electrical interfaces for soft electronics.[145] In addition, another consideration with liquid metal is cost, which is higher (~$1/g) relative to the other materials in this review.

3.2. Carbon-Based Conductors

Due to their electrical conductivity, ease of processing, and elemental abundance, carbon allotropes such as carbon nanotubes (CNTs) and graphene have been studied for applications in stretchable electronics.[146,147] Although individual particles of these carbonaceous materials are inextensible, they can be deposited as thin percolated films on elastomeric substrate that allow them to have useful electromechanical properties. With addition of surfactants they can be dispersed in a variety of solvents that makes them suitable for printing.[148,149] After dispensing onto a surface, the solvent evaporates, leaving behind a connected network of conductive particles. Upon stretching of the underlying substrate, the conductive particles can rotate, flex (straighten), and slide/slip to maintain electrical contact.

The superlative properties of individual 1D[154] and 2D[155] carbon structures are challenging to replicate for large area and printed electronics.[156] For example single or a few CNTs are known to have conductivity in the range 10^4−10^5 S/cm.[157] Whereas, thin films (100−400 nm thickness) of such 1D structures show percolative electrical transport which typically show conductivity in the order of 10^2 S/cm.[150] Therefore, it is desirable to distribute the particles uniformly during printing to avoid agglomerates. For instance, the use of surfactants to disperse CNTs for inkjet printing results in a film with lower sheet resistance compared to that of CNTs printed without surfactant. Postprocessing methods, such as acid treatment of the printed structures, can decrease the amount of surfactant present in the printed film, thus, lowering the sheet resistance (Figure 8a).[150] Electrical conductivity can also be tuned by varying the thickness in such films which can be ascribed to improved percolation paths to conduct charge.[150,158] The acid treatment, if performed before printing CNT as a purification step, results in a stable dispersion without any surfactant.[159] This approach is particularly useful when post-treatment can adversely affect the device performance (Figure 8b). For inkjet printing of CNTs in particular, it is important to filter the ink before printing as the CNTs with large aspect ratio can lead to clogging of the nozzle.[160] As general rule, it is suggested to keep the length less than 1/50 of the nozzle diameter.[161,162]

Once filtered the dispersion should also remain stable over time to prevent agglomeration of CNTs. The stability of dispersion can be quantified using a zeta potential analyzer with higher value indicating a stable dispersion due to repulsion between CNTs. Although, a high boiling point solvent is preferred to disperse CNTs to mitigate the coffee ring effect upon printing, addition of solvents such as isopropyl alcohol is useful reducing the surface tension. A lower surface tension ink readily wets the inkjet nozzle and plasma treated elastomer (plasma treatment leads to improved adhesion even under

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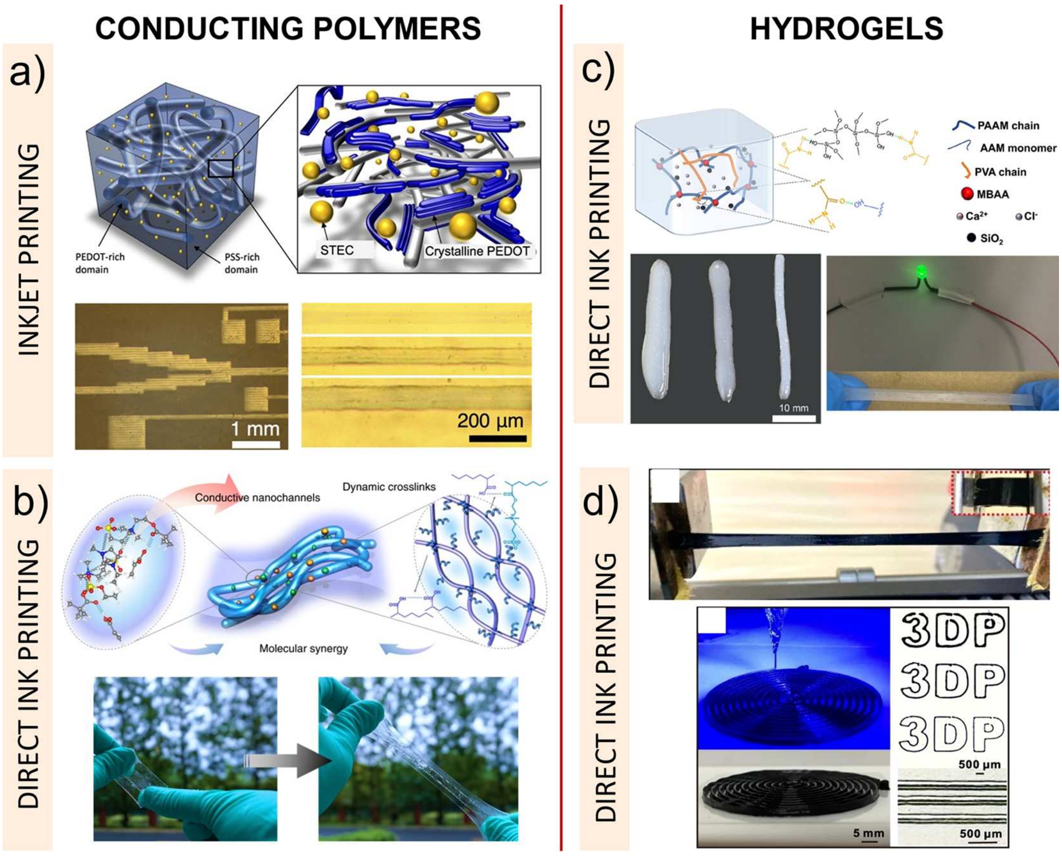

Figure 9. Printing stretchable conducting polymers. a) Addition of ionic additives to PEDOT:PSS results in increased conductivity and stretchability. The modified PEDOT:PSS can be inkjet printed with a minimum line width of ≈40 μm. Reproduced with permission.[41] Copyright 2017, American Association for the Advancement of Science. b) A polyzwitterion, acrylic acid, and ionic liquid form a stretchable conductor that can be inkjet printed. The conductivity results from nanochannels in the material and the mechanical properties arise from dynamic cross-links. The extruded polymer film is transparent in visible region, stretchable, and conductive. Reproduced with permission.[184] Copyright 2019, Springer Nature. Printing conductive hydrogels. c) Poly(acrylamide) (PAAm)-*co*-poly(vinyl alcohol) hydrogels extrusion printed with varying fraction of fumed silica particles that act as a rheological modifier. Reproduced with permission.[188] Copyright 2022, Wiley-VCH. d) Starting with the monomer EDOT instead of the polymer PEDOT can lead to improved structural integrity as seen from the reduced spreading the printed filaments (left to right). Careful selection of photoinitiator and cross-linker can decrease the photogelatin time to enable high resolution extrusion printed stretchable conductors. Reproduced with permission.[189] Copyright 2021, Springer Nature.

strain[163]). Once the wettability of the ink is tuned, the final step is to optimize the printing parameters such as droplet spacing and substrate heating. For example, in one study, the space between two consecutive droplets and the extent of droplet spreading determines continuity and the morphology of the print. The substrate temperature helps in decreasing the droplet spread via faster solvent evaporation. For example, in a study on inkjet printing of CNT dispersion (IPA as a solvent), it was found that a droplet spacing of 127 μm and a substrate temperature of 60 °C resulted in the most optimally printed CNT tracks.[161]

Printing exfoliated graphene sheets for stretchable conductor or interconnects is challenging due to crack formation in the deposited film at low strain values (<5%) since graphene sheets cannot easily slide past each other during stretching of the substrate.[164,165] Using stretchable textiles as a template to print graphene is an attractive strategy to overcome this challenge. By adjusting print parameters such as number of passes and ink concentration, graphene can conformally coat textile threads (Figure 8c). However, despite the use of a textile substrate, cracks start appearing in the films after multiple cycles of 25% strain.[152] Another approach to facilitate stretchability in graphene is to combine hierarchical structures from the microscale to macroscale. At the microscale, a combination of graphene and CNT contribute to the strength by increasing mutual van der Waals interaction mimicking reinforced rebars (Figure 8d). The CNT-graphene reinforced structure at microscale combined with an extrusion printed mesh like structure at a macroscale, results in high stretchability (200% strain).[153]

Other than the printing approaches discussed here, laser writing is another commonly used strategy to pattern carbon-based materials, especially graphene. The process typically involves a scanning a CO_2 infrared laser on polymer like polyimide (PI)[166] and SU-8[167] that results in carbonization of the polymer. The resultant graphene structure is commonly referred to as laser-induced graphene. The patterned regions can then be selectively transferred onto a stretchable substrate to be used as a stretchable conductor. Further details about the process and material selection can be found in this focused review.[168]

Although, studies on printed stretchable conductors using carbon-based materials are scarce, they are promising materials for stretchable electronics. A major advantage of using graphene and CNTs is their environmental stability. When compared, metal nanostructures such as AgNWs/NPs have superior

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electrical conductivity, but they get oxidized over time that lowers their electrical conductivity. Although, this problem can be solved by passivating the surfaces,[169] but it adds another step in the process, and an additional interface which can limit their electromechanical performance. In addition, both graphene and CNTs are promising electrode materials for electrochemical devices. This ability combined with their mechanical properties[170−172] has led to applications such as stretchable supercapacitors and batteries.[173,174]

3.3. Conducting Polymers and Gels

Conducting Polymers. Conducting polymers show electrically conducting or semiconducting behavior due to their conjugated structure.[175,176] One of the promises of organic electronics is that the conductors can be synthesized, dissolved in solvent, and deposited or printed onto surfaces at ambient conditions. While there is a large body of literature on conducting polymers, inclusion in this review requires the material to be both printable and stretchable, which significantly lessens the range of materials that qualify. In general, conducting polymers are not inherently elastic. For example, while PEDOT:PSS[177,178] is one of the most studied conducting polymers for electronic applications, it undergoes plastic deformation when strained and fails within a strain of 10%, limiting its application in stretchable electronics.[178] To address these issues, researchers have used either additives or flexible segments to soften conductive polymers (albeit at the expense of conductivity) or by using phase behavior to get conductive domains to coexist among elastomeric networks, such as in blends.

Adding a plasticizer, such as Triton[179] can soften PEDOT:PSS. The plasticizer gives the material viscoelastic, dough-like behavior. The plasticizer coerces PEDOT:PSS to phase separate into PEDOT nanofibrils within the dough, which is suggested to be the reason for an improved electrical conduction compared to the pristine PEDOT:PSS.[180] The nanofibrils domains are not inherently stretchable, but due to their meandering path within the plasticized matrix, the resulting material effectively becomes deformable (e.g., the fibrils can extend/straighten while maintaining conductivity). Further increase in the concentration of plasticizer decreases the electrical conductivity, but increases stretchability, indicating a trade-off. The PEDOT:PSS dough can be readily screen printed on PDMS substrates and can sustain up to 50% strain while maintaining an electrical conductivity of ∼10 S/cm.[11] After stretching, the dough slowly recovers (∼30 s to return to zero strain after stretching to 30% strain).

Addition of ionic liquid can also improve the stretchability and conductivity of PEDOT:PSS. Ionic liquids have a charge screening effect that weakens the interaction between PEDOT and PSS domains. The PEDOT nanofibrils form an interconnected network with improved crystallinity that increases conductivity compared to the pristine PEDOT:PSS. In addition, the ionic liquid further softens the PSS matrix making the polymer more stretchable. These type of copolymers with hard and soft segments are commonly found in thermoplastic elastomers.[181] The PEDOT:PSS dispersion with ionic additives can be inkjet printed on an elastomer with a resolution of 40 μm (Figure 9a). The electrical conductivity of the printed structures (3600 S/cm) remain constant even at 100% strain.[182]

Polyzwitterions—that is, polymers with both cationic and anionic charge—are a class of polymers that can conduct.[183] When mixed with an ionic liquid and copolymerized with acrylic acid, they are capable of forming dynamic hydrogen bonds along the backbone of the polymer (Figure 9b). These bonds improve the mechanical properties while the formation of so-called "nanochannels" helps provide electrical conductivity.[184] However, unlike PEDOT:PSS the polyzwitterions do not have crystalline conductive domains and therefore, the electrical conductivity is not as high. The ink made from polyzwitterion solution shows shear thinning behavior and can be extruded for direct-write printing. The printed material has strain invariant conductivity up to large strains (1000% strain) but shows poor conductivity (10^{-4} S/cm).[182]

Conducting polymers can also be patterned by methods other than printing such as optical lithography. One concern of using lithography for patterning is the acidic nature of PEDOT:PSS that can interfere with the chemistry of conventional photoresist.[185] Other methods of patterning include electro-polymerization[186] and selective wetting,[187] although these approaches have not been used to create stretchable conducting polymers

Hydrogels. Hydrogels are hydrophilic cross-linked polymer networks with large water content. Printing hydrogel has been a topic of research for more than a decade, with the initial focus on scaffold printing for artificial organ or tissues.[190,191] Hydrogels can be elastic (stretchable) and despite having modest conductivity via ionic mechanisms (10^{-1}–10^{1} S/cm), they have gained interest recently for "ionotronics", which are devices that have some analogy with electronics but use ions instead of electrons.[192] Such ionotronic devices are motivated by the properties imparted by hydrogels including optical transparency, bio- and enviro-compatibility, and ionic functionality; this latter attribute is particularly interesting since the body also conducts and functions using ions.

One way to print hydrogels is to print them as liquid monomers or oligomeric precursors that solidify by photopolymerization during- or postextrusion. A major factor that affects the process of printing is time taken to polymerize and cross-link, which is referred to as curing time or gelation time. This is important especially when multiple printing passes are needed. The curing time can be tuned by varying the choice of monomers/oligomers, molecular weight and concentration of the cross-linker, and water content.[193] These parameters are also used to tune the stiffness and stretchability of the hydrogels.[193,194] Soaking the hydrogels in salt solution or dissolving salt into the prepolymer during synthesis are the two common approaches for making them conductive. When the gels are cured postextrusion, the rheology of the pregel should be tuned so that the print resolution is not compromised. Adding fillers like fumed silica particles help in rheological modifications for better print resolution. Further, functional groups on the fumed silica particles can become additional cross-linking sites, thus, improving the mechanical strength of the hydrogels.[188] The effects of fumed silica particles (labeled as SiO_2 in Figure 9c) leading to additional cross-linking sites and improved print resolution are shown in Figure 9c.

Hydrogels can contain conducting polymers that offer mixed ionic-electronic conduction. These are particularly useful for applications as a stretchable conductor for low-impedance interfacing electronic systems with biological tissues and organs.[195,196] As a limitation, conducting polymers like PEDOT:PSS can readily absorb and thereby block the incident radiation needed for photopolymerization, which hampers the gelation of the sample. Using a monomer (EDOT) instead of polymer (PEDOT) and a photoinitiator helps resolve this issue.

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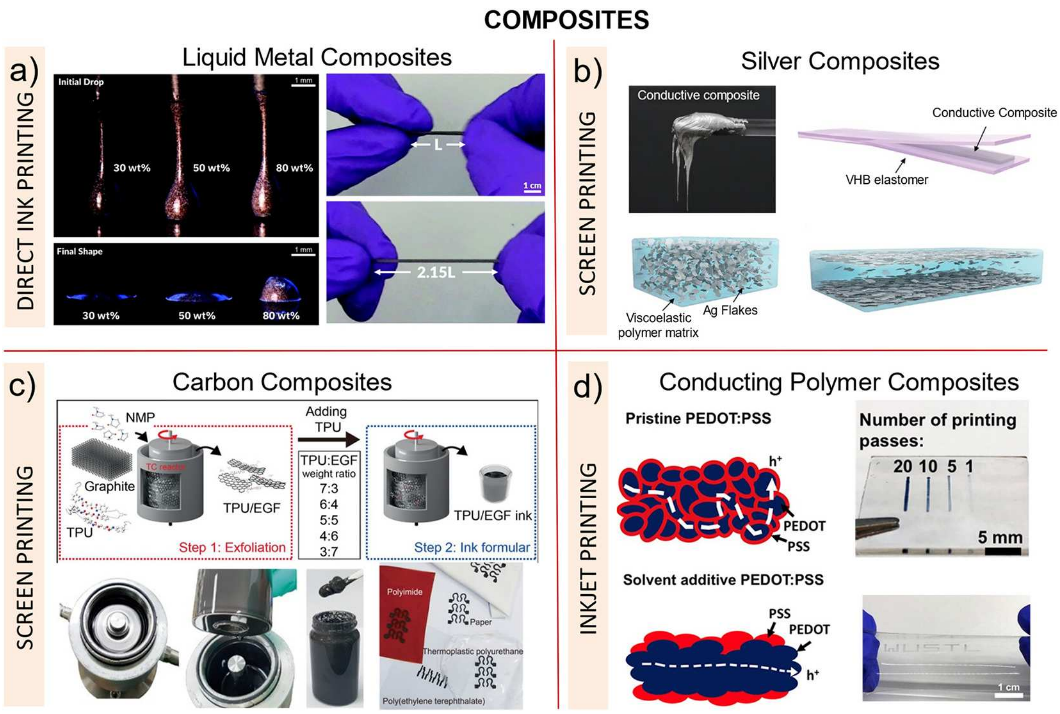

Figure 10. Composite materials for printing stretchable conductors. a) Bulk liquid metal can be dispersed as particles in silicone by simple mixing. Increasing the liquid metal particle concentration imparts shear thinning behavior to the composite which is useful for direct ink writing. Reproduced with permission.[212] Copyright 2020, Royal Society of Chemistry. b) Ag flake composite. Viscoelastic nature of the composite allows a redistribution of Ag flakes within the polymer matrix under strain. This redistribution results in strain invariant electrical conductivity. Reproduced with permission.[43] Copyright 2022, Wiley-VCH. c) Fluid dynamic based exfoliation of graphite improves the yield and the dispersibility of graphene sheets in thermoplastic polyurethane (TPU). A photograph of the reactor for graphite exfoliation and the resultant graphene-TPU composite ink. The graphene-TPU composite inks can be screen printed to make a stretchable electrode. Reproduced with permission.[40] Copyright 2021, Wiley-VCH. d) Adding poly(ethylene oxide) (PEO) to PEDOT: PSS improves its stretchability and ethylene glycol (EG) a polar solvent improves its electrical conductivity. Reprinted with permission.[214] Copyright 2021, American Chemical Society.

The extrusion-printed structures have short gelation time, high stretchability and show a small change in resistance up to 100% strain (Figure 9d).[189]

The limitations posed by gelation time can be overcome by "embedded printing" of conductive hydrogel in a support matrix. As an example, the support matrix may consist of a physically cross-linked gel. In the first step to make the support matrix, the polymer (alginate) forms ionic cross-links with Ca^{2+} ions. Further processing in the next step results in the formation of a granular fluid with a yield stress. This granular fluid is the support matrix. A conductive hydrogel ink is prepared using Ag flakes. Due to shear thinning behavior of this yield stress fluid, the conductive hydrogel ink can be readily extruded in the supporting matrix without letting the gravity affect the structural integrity of the printed filament. Once printed, the matrix with the embedded filament can be thermally cured to allow chemical cross-linking and the printed part remains in the support matrix.[197]

There are many other methods to pattern hydrogels. The most facile approach being soft lithography or molds with desired geometries. Patterning methods that use lasers to pattern hydrogels are stereolithography (UV laser) and two-photon polymerization (femtosecond IR lasers are used in general).[198] Among the two, the two-photon polymerization has gained a lot of traction due to its ability to generate 3D features with submicron resolution.[199] Besides these methods, hydrogels can also be patterned using UV light sources and shadow masks to localize cross-linking in the gel.

3.4. Composites

In this section, we focus on composite inks comprised of conductive and nonconductive phases. Typically—but not always - the conductive phase consists of stiff materials that are not inherently stretchable, but when distributed and percolated in a nonconductive, elastomeric phase, can create stretchable conductors. Liquid metals—either as a filler or as a continuous phase—are the obvious exception. Regardless of the filler, charge transport in such composites depend on the distribution of conductive fillers and is described by percolation theory.[200] A variety of materials are used as conducting fillers such as liquid metal, metal nanostructures (nanoparticles, nanowires and nanosheets), carbonaceous materials, and conducting polymers. Generally, elastomers such as silicones are used as nonconductive matrices. For more information on such materials, there are focused reviews in the literature.[201,202] Here, composites are categorized on the basis of four different types of conductive fillers which are commonly studied for printed stretchable electronics.

3.4.1. Liquid Metal Composites. Printing liquid metal is challenging mainly due to its large surface tension and low viscosity; simply stated, applying enough pressure to overcome the surface tension causes liquid to rapidly come out of a nozzle due to the low viscosity. The section on liquid metal discusses one way to overcome this issue by using adhesion between the oxide and substrate to enable printing through the use of shear or tension. Alternatively, the rheological properties of the metal can be altered. Ideally, doing so would allow the metal to be extruded from a nozzle as a filament rather than a droplet or blob. Adding particles is one way to modify the rheological behavior of liquid metal. Composite liquid metal inks with metal[203,204] or nonmetal[205,206] particles can be obtained by sonicating,[50] stirring,[203] ball milling,[205] or mixing with mortar and pestle the particles in the presence of the liquid metal.[207] In these inks, the liquid metal is the continuous phase. Due to large interfacial tension of liquid metal,[129] it is challenging to mix nonmetallic additives with liquid metal. However, the oxide that forms on liquid metal can help “engulf” particles by stirring the particles with the metal in the presence of air.[208] Metal particles, such as Cu, can mix directly with liquid metals in the absence of oxide via the formation of metal−metal bonds, but are known to form intermetallic phases with liquid metal, which can lead to time dependent variation in rheological behavior.[207] Nonmetal additives such as carbonaceous materials[209] or quartz particles[205] impart viscous, paste like behavior to liquid metal which is useful for printing, but the electrical conductivity is compromised to a certain extent. One challenge with such composites is to prevent particles from jamming at the nozzle during extrusion printing.

Dispersing liquid metal droplets or particles in elastomers (i.e., elastomer as the continuous phase) offers an alternate route to make composite inks that automatically contain an elastomeric encapsulation layer to protect the printed structure. For more information readers can refer to focused reviews on this topic.[210,211] Addition of liquid metal particles increases the viscosity of elastomers, imparting a shear thinning rheology which makes it easy to extrude and helps retains the shape once printed (Figure 10a).[212] Due to the low aspect ratio of liquid metal particles, they do not readily percolate within the elastomeric matrix. The liquid metal particles in the elastomer matrix needs to be sintered/coalesced postprinting to make the composite electrically conducting (of the order: 10^3 S/cm). This can be done either by applying shear[212] or acoustic[209] force to rupture the thin layer of oxide on the metal and any polymer between the metallic particles. Alternatively, other particles can be mixed with the liquid metal, such as silver, to produce an interconnected metal network in elastomer; in the case of silver with gallium indium, the particles alloy to form solid AgIn particles connected by liquid GaIn to form a highly conductive composite (of the order: 10^4 S/cm).[213]

3.4.2. Silver Composites. Silver is perhaps the most studied material for making stretchable conductive composites due to its high conductivity, availability (flakes are commercially available and Ag particles are easy to make by reducing Ag+ salts), and its slow tendency to oxidize. As with other composites, it is important to maximize percolation between particles to maximize conductivity (Ag composites have conductivities ranging from 10^2 to 10^4 S/cm). Although, noble metals that lack oxides such as Pt and Au would be preferable for making conductive contacts, they are more expensive than Ag. In the case of nonstretchable Ag conductors, conductivity can be improved by sintering to help make better contact between particles (e.g., by removing ligands or surface oxides). Thermal sintering only works if the underlying substrate is compatible with elevated temperatures. Alternatively, light assisted sintering is a viable approach because it delivers heat briefly to the particles using an intense flash of light.[215,216] Although sintering is typically not used to create stretchable Ag composites (since fused particles are not extensible), there is at least one example where sintering was used to make clusters of Ag particles embedded in an elastomeric matrix.[217] Instead of sintering, another approach to promote particle−particle contact is to mix additives (such as the surfactant, Zonyl) into the elastomeric composites that help promote contact between Ag particles.

Composites make use of different structural forms of silver such as flakes,[218,219] nanowires[220] and nanoparticles[72] with each having its own advantage. A key consideration is maintaining contact between the solid Ag particles while deforming the encasing elastomer. In a composite, vertically stacked flakes or 2-D structures help in maintaining electrical conductivity with minimal electrical fluctuations during straining by sliding action.[218] High aspect ratio structures help improve the conductivity (for a given mass loading of Ag) by forming better percolated networks.[220,221] Nanoparticles have the ability to move (rotate, slide) to preserve the percolating pathways that get distorted under strain.[18,222] Combining two different Ag particle morphologies further helps in improving the electrical and mechanical compliance under strain.[18,223] Another strategy to tune the performance is varying the volume fraction of silver fillers and additives like phosphotungstic acid (PTA) that changes the viscoelastic behavior of the composite before and after printing. Additives like PTA can form dynamic hydrogen bonds with the polymer matrix to allow spatial movement or reorganization of Ag flakes in the matrix under strain. This reorganization prevents loss of electrical conductivity even at strain values as high as 1000%. To promote such a reorganization of conductive fillers, it is desired to have a storage modulus larger than loss modulus for lower strain, and a reverse relation for larger strain values (Figure 10b).[43] This viscoelastic behavior is suitable for certain printing techniques such as screen printing.[43,220] Depending on the type of printing, different set of functionalities are required in the composite ink.

For reverse offset printing, a semidry film with sufficient cohesive strength is needed for reliable transfer of pattern from an ink-coated roll to the substrate. A combination of a low and a high boiling point solvents maintains the semidry state, while binders like polyacrylate ensure sufficient cohesion.[223] For direct ink printing, the viscosity of the composite ink can be tuned by adding a lubricant, which can improve the dispersibility of Ag filler, leading to improved conductivity under strain.[224]

3.4.3. Carbon Composites. Compared to metals, which are prone to oxidize, carbon based fillers are chemically more stable[225] which makes them particularly useful for printing stretchable electrodes that can be used in batteries[226−228] and wearable sensors.[229] Carbonaceous materials such as graphene and carbon nanotube are also used to augment the electrical and mechanical behavior in metal-based conductive composites.[230−232] Carbon composites typically have conductivity ranging from 1 to 20 S/cm.

Graphene is available as stacked sheets in graphite, but the key challenge is to separate the sheets into a suspension. Thus, to make graphene composites, the first step is to synthesize graphene oxide since pristine graphene itself is difficult to disperse in polar or nonpolar solvents without a surfactant because it is hydrophobic. Graphene oxide is commonly created by liquid exfoliation of graphite, followed by mixing with an elastomer. Liquid exfoliation of graphite is a multistep process with a very low yield.[233,234] An alternative, fluid-dynamics or fluidic-delamination process combines multiple forces−shear, pressure and collision - to exfoliate graphite into high quality graphene oxide sheets. In the second step, mixing the exfoliated graphene oxide flakes with elastomer in the presence of a solvent produces a paste, as shown in Figure 10c.[40] The solvent can be evaporated pre- or postprinting to tune the rheology of the

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composite ink as per the printing method. The graphene composite ink can be screen printed on a variety of substrates (thermoplastic urethane, polyethylene, and polyimide, as shown in Figure 10c) resulting in conductors with high stretchability (up to ~300%) without significant change in conductivity (4.33 S/cm).[40]

Carbon nanotubes (CNTs) show poor dispersibility in silicone elastomers, resulting in its aggregation. The dispersibility of CNTs in silicone can be improved by selecting a solvent with a comparable Hansen solubility parameter to that of silicone (e.g., *n*-butyl acetate). Homogenous dispersion also prevents nozzle clogging during direct write printing. By suitable functionalization of CNTs these composite inks can be printed on any desired substrate.[235] Once printed, humidity sensitive silicones like RTV (room temperature vulcanizing) makes the process of curing easier compared to the commonly used two part silicone elastomer.[235]

3.4.4. Conducting Polymer Composites. Most of the printed conducting polymers reported so far have used PEDOT:PSS as a conductive filler in an elastomeric or hydrogel matrix. Their conductivities are typically in the range of 30−120 S/cm.[214,236] The latter imparts desirable mechanical properties, while the former provides electrical conductivity. Choosing the right solvent or a combination of solvent is important, as it affects the print quality and conductivity.

Even after addition of surfactants, the coffee stain effect is observed in inkjet printed PEDOT:PSS composites.[237] Using a higher boiling point cosolvent (e.g., ethylene glycol) that has lower surface tension than the primary solvent (e.g., water) results in Marangoni flow, that counter balances the convective flow of the composite ink.[237,238] This results in uniform distribution of solute in the printed features. Polar solvents like ethylene glycol play a dual role by also improving the electrical conductivity of the PEDOT:PSS. Water-soluble polymers like poly(ethylene oxide) (PEO) and thermoplastic urethane (TPU) can form blends with PEDOT:PSS to impart stretchability.[214,237] To improve the electrical conductivity of PEDOT:PSS−PEO blends, ethylene glycol can be used since it is a polar solvent. It is hypothesized that the addition of ethylene glycol and subsequent annealing (120 °C) results in separation of PEDOT and PSS phases which improves the percolative transport (Figure 10d). The electrical conductivity of PEDOT:PSS blends can also be improved by printing multiple passes which results in thicker traces, as shown in Figure 10d. Miscibility of hydrophobic elastomers such as PDMS with PEDOT:PSS is a problem that can be solved by using surfactants like Triton X-100.[239] The addition of Triton X-100 also results in phase separation of PEDOT and PSS regions that crystallizes the PEDOT phase, improving the electrical conductivity.[236,240] Use of amphiphilic polymers like PDMS-*b*-PEO is an alternate way to make composites with PEDOT:PSS (Table 1).[241]

4. DISCUSSION

This guide discusses printing methods and inks to produce stretchable conductors. Comparing different printing techniques is nontrivial because there are so many considerations. Printing comes in different shapes and sizes, literally. From the large industrial scale printers to the smaller hobbyist-grade ones, each has its own "best practice" and governing principles, not to mention the less documented know-how privy to those who are already proficient at the technique. In addition, more conductive inks and stretchable composite structures are rapidly emerging in the literature.

The array of theoretically compatible combinations of printing technique and stretchable conductive ink is vast. While this space presents exciting opportunities for innovation, it is also overwhelming to be faced with so many options. Ultimately, to an engineer searching for a means to an end, the crux of the matter is "Out of the available options, what is the most appropriate process and material(s) to achieve said goal?" We hope that this article has been helpful in providing guidance for such a decision-making process.

To reiterate, often the foremost step toward a suitable procedure to fabricate a certain stretchable conductive object is a well-defined intention, since the design requirement informs the processing technique and choice of materials. For example, if mass production of a certain design with fine features is desired, gravure printing is an attractive option, and committing to this particular technique immediately rules out groups of materials (e.g., bulk liquid metal—high surface tension and unique rheology presents undesirable dewetting complications). Utilizing Ashby charts is a good start for materials selection, which we have demonstrated here (cf Figure 2). Using a comprehensive materials database (e.g., Ansys Granta Selector), one can create charts with different material properties and other factors like cost. In short, adopting a systematic approach, such as a process of elimination centered around a clear goal, helps to overcome design-related decision fatigue.

5. CHALLENGES

Although, there have been many advances in the field of printed stretchable conductors, there are some challenges that need to be addressed. We have identified four major areas.

5.1. Printability

There are several reports on the development of new materials inks for printing stretchable conductors, but few focus on rationalizing them based on their properties. One of the important properties is the printability of the ink. Studies that focus on parametrizing the properties such viscosity and surface tension, that govern printability for a given combination of material and printing method, will be very useful. For example, rheological studies helped in defining a "printing zone" or printability for extrusion printing of liquid metal paste.[251] Similarly, in another study peak hold test was used to mimic the conditions during screen printing or extrusion printing.[252] Another challenge concerns with the use of inks based on nanomaterials. When printing nanomaterials like AgNWs and CNTs large aspect ratios are useful for improving the conductivity. However, large aspect ratios also result in nozzle clogging indicating a trade-off. Standardizing the parameters for printing a material ink will go a long way toward advancing the field and ultimately commercialization.

5.2. Reliability

Printing stretchable conductors with reproducible shape and electromechanical performance remains a challenge. Uniformity of printed structures across multiple printing passes would reduce any geometry induced variation in electrical conductivities across different samples. This aspect of uniform print geometry gains more importance in realizing high-density (<10 μm gap width) printed stretchable transistors. Here, slight variations in print geometry could lead to shorting of interconnects or devices. To address this challenge, parameters for each printing method and the ink needs to be optimized on a case-by-case basis. In case of inkjet printing, for instance, ensuring uniform droplet generation, understanding the droplet

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drying behavior and droplet coalescing behavior can lead to significant improvement in print uniformity.[86,253]

The printed conductors should also have reproducible electromechanical behavior. This implies ideally zero hysteresis across multiple strain cycles. More practically, it would translate to hysteresis with permissible tolerance defined by the application. Although, there are no industry standards for this, the community can direct efforts in establishing reliability data. These studies should be designed in a way that they involve different modes of deformation (bend, stretch, torsion and twist) to capture realistic conditions.[254] The electromechanical behavior can also get affected by constant current induced material degradation, but has been scarcely studied.[255] An important parameter, device lifetime can be deduced from such studies.

In addition to electromechanical reliability, any commercial applications will have to survive rigorous environmental testing before making it to market. Devices need to maintain functionality across a wide range of temperatures in addition to surviving accelerated testing at elevated temperatures and relative humidity.

5.3. Printing Hardware Reusability

A major challenge in printing that is seldom addressed is the reusability of the printing hardware used in the research laboratories. This is important as it not only affects the print resolution but also affects the overall cost and throughput of printing. The reusability aspect can be understood on a case-by-case basis for each printing method.

For nozzle-based printing method, clogging is a common problem. How does one address this issue on the equipment side? How does one estimate the dispensed volume after which nozzle needs to be cleaned or replaced? What are the methods physical or chemical to clean the nozzle and render it suitable for reuse? The answer to these questions are specific to the ink type and they are a function of parameters such as but not limited to particle size, printing speed, nozzle diameter and preprinting steps like in-built filtering in nozzle.[83]

In the case of roll printing method such as gravure printing, some of the major tool related challenges are residual ink in the cell, and doctor blade wear. Continuous use of the printer can also lead to ink drying along the cell walls. The problem of residual ink can be resolved by designing cells with suitable aspect ratios.[256] Solvent drying can be avoided by using higher boiling point solvents or by keeping the instrument in controlled environment.[37] Worn doctor blades need to be replaced.

For reverse offset printing, another roll printing method, the main tool issues are related to the cliché and PDMS roll. The cliché needs to be cleaned periodically to remove residual ink, while the PDMS roll needs to be monitored for any major deformation because of continuous contact with the cliché. The PDMS roll plays an important role of absorbing excess solvent and rendering the film in a desired semidry state. However, after multiple printing passes the absorption ability of PDMS slowly decreases which results in nonuniform print patterns.[111] A rigorous analysis of these and other process parameters will certainly help in optimizing the process and improving the reproducibility of the print.

5.4. Resolution and Throughput

High-throughput, high-resolution (<10 μm) printing of stretchable conductors can lead to a new generation of high-density stretchable electronic devices. Currently, there is a trade-off between high resolution and throughput: achieving both remains a major challenge. Although roll printing methods such as gravure and reverse offset printing offer a promising approach to address this challenge, the patterns cannot be changed on the fly as they can with nozzle-based printing methods. Roll printing techniques also require materials with specific rheological properties; thus, not all materials are compatible with roll printing. Material selection and patterning to make cliché in reverse offset or ink carrying metal cells in gravure can overrun their advantage of printing high-resolution features at large scale.[37] Furthermore, roll techniques require the fabrication of the "roll" itself. Conventional methods including laser engraving fail to provide resolution beyond 10 μm, whereafter, cells for high-resolution printing are made from silicon using micro-fabrication methods.[37] A similar argument can be made for screen printing where improving print resolution beyond 50 μm requires use of silicon stencils, that are made using multistep and time-consuming microfabrication techniques.

Nozzle-based printing methods such as inkjet and extrusion printing can achieve high throughput using multinozzle systems, but there is a major challenge in achieving spatial precision. Another area of focus to improve the throughput for these methods is the development of new algorithms for toolpath optimization. This becomes even more important in the case of continuous extrusion where the material flow cannot be stopped or paused.[257] An optimized toolpath would reduce the material wastage and ensure uniform printing. The challenge of improving resolution in nozzle-based printing is a two-part problem: materials and methods need innovation. Extruding conductive composites inks through thinner nozzles (diameter <150 μm) becomes challenging due to nozzle clogging.[258] Although, reducing extrusion speed can improve issues associated with clogging, it adversely affects the throughput. Using a combination of phase-change materials and nozzle heating to a suitable temperature, desired extrusion rate can be maintained without clogging the nozzle.[258] However, die swelling (i.e., the expansion of the printed filament as it exits the nozzle) can still compromise the resolution.

6. FUTURE TRENDS AND TECHNOLOGIES

Innovations in printed stretchable conductors could transform the way large area unconventional electronic devices are manufactured. Several examples are provided below:

6.1. Multilayer-Multimaterial Printing

Efforts from researchers have focused on material and printing method innovations to improve the performance of standalone printed stretchable conductors. To enable their true potential, it is important to seamlessly integrate printed stretchable conductors with individual components and eventually a system with desired functionalities. In this regard, we envisage that multilayered-multimaterial printing will play a key role and offer multiple areas for innovation. One such area is printed stretchable vertical interconnect access (VIAs) that provide reliable interlayer connectivity under mechanical deformation. These interconnects can lead to robust multilayered stretchable electronic devices.[259,260] Another area of research includes material design and process planning/modification to ensure chemical and thermal compatibility of printed materials. This consideration is to ensure that chemical or thermal treatment of one layer (e.g., thermal sintering of Ag NWs) should not affect the performance of preceding layer (e.g., hydrogel). Multilayer-multimaterial printing also warrants study of wetting behavior of each material on the target layer (substrate). This will not only

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help improve conductor-substrate adhesion across multiple layers, but also prevent materials mixing when two different material inks are printed adjacent to each other.[261] For more information, readers are directed to a focused review on this topic.[262]

6.2. System Integration

Combining computational power of conventional electronics with stretchable conductors can lead to many exciting applications across multiple domains. To enable conformability, this approach would typically require integration of thinned Si integrated circuits (ICs) with printed components. A major challenge here is the mismatch in the stiffness of rigid ICs and soft printed conductors. Developing strategies to design and print conductive materials with variable stiffness offers a promising solution to this challenge and an exciting avenue for research.

Another aspect of system integration is interfacing with external hardware, that would require intimate electrical contact of stretchable conductors typically with flexible flat cables (FFC) at one end. Developing printable and stretchable material inks that also demonstrate adhesive behavior behavior can be useful in this regard. Such materials can also be used to adhere rigid Si ICs onto a stretchable substrate, removing the need for using an additional conductive adhesive that could result in multiple interfaces.

6.3. Self-Healing Stretchable Conductors

Stretchable electronics experience mechanical stress during their use. Thus, the ability to self-heal can improve the reliability of printed stretchable conductors. Although there is extensive literature on self-healing conductive materials,[263] there are very few examples of such materials being integrated with printing processes.

6.4. Stretchable Fibers

Electrically conducting stretchable fibers have been recently studied for applications in smart textiles,[264] wearable,[265] and implantable[266] electronic devices. The ability to print these individual stretchable fibers with desired functionality, or printing materials on commercially available textiles to impart them with desired functionalities, can lead to several exciting applications.[267]

6.5. Emerging Printing Methods

Embedded 3D printing represents one of the recent developments in printing stretchable conductors (see section 3.1 for details). An advantage of this method is that it eliminates the problem of delamination caused due to layer-by-layer patterning.[268,269] Another variation of direct printing involves use of magnetic field (direct magnetic printing). So far, this method has been predominantly used for printing inks made from liquid metal mixed with magnetic particles (e.g., Ni).[270−272] The advantage of this method is its facile setup, and recently it was shown that its resolution can be tuned (down to 100 μm) by magnetic printing in laser defined features.[272]

Meniscus guided printing is another method that has been successfully shown to print stretchable conductive filaments with a resolution of 50 μm using liquid metal particle based composite inks.[273] The basic underlying principle in this method is that the solution meniscus forms an air−liquid interface that results in solute deposition as solvent evaporates.[274,275] When used in direct printing, the meniscus forms between the tip of the moving nozzle and the substrate.[273] Although, this method is promising due to high resolution (50 μm) and high print speed (6 mm/s), the ink design is challenging. Several factors such as rheology, solvent evaporation, and effect of solute concentration needs to considered to ensure crack free continuous printing.[275]

Another interesting development is integrating existing printing methods with a robotic arm to achieve a high degree of spatial freedom beyond that of a standard 3 axis stage control. Such integration has been reported for printing soft neuromorphic devices[276] and multimaterial electronic devices,[277] and making them stretchable for soft robotics or electronic skins is an interesting proposition.

CONCLUSION

Printing stretchable conductors is an attractive approach toward the development next generation soft electronic devices. The biggest advantage of this approach is the versatility offered by the "tool kit" of printing methods and materials. This versatility can enable applications including high density stretchable transistors, biomedical devices that have tissue like softness and robust electrodes for wearable batteries. Printing is also attractive because it is a form of additive manufacturing in which material is placed only where it is needed and often in a way that lends itself to rapid prototyping. This approach of printing also offers several avenues for innovation such as optimization of existing printing methods to push beyond their limits or development new methods. Similarly, with materials there is a large palette to explore.

This review aims to guide the selection of a printing method and a material for printable, stretchable conductors. The material selection is based on parameters like viscosity, resolution, electrical conductivity, and stretchability. We then classified printing methods and materials. While discussing them in detail, we identified their limitations and suggested strategies to overcome them. Finally, we highlighted major challenges and identified future trends and technologies that could drive innovation in this field.

To achieve the complete potential offered by printing stretchable conductors, it is important to identify and solve the fundamental issues in the field. The electromechanical reliability over thousands of deformation cycles is one such issue that is seldom studied or reported, but is crucial for any practical application. Similarly, in many studies the inks are developed without systematically studying their rheology for a particular printing method and application. The field would improve by future studies rigorously analyzing the properties of material inks for each printing method and application. This can be done for example by identifying suitable rheological properties of the ink (viscosity, yield stress, storage modulus), wetting behavior (contact angle), surface tension, and printing parameters (e.g., print speed, gap height and nozzle diameter for direct ink printing). Another aspect of reliability is environmental stability of printed conductors. Several conducting materials such as hydrogels or metal nanostructures are prone to environmental degradation. This offers an opportunity for researchers to develop strategies to protect these materials that can be preferably be integrated with the printing process.

On the equipment side, off-the shelf material printers are not tailored for every possible material ink, and sometimes the modifications to the equipment need to be made locally by the researchers in the lab. We suggested instances in which existing printing equipment was modified to overcome the limitations. One such example discussed was the use of an acid impregnated filter at the nozzle tip of an inkjet printer to prevent oxide formation in LM, that clogs the nozzle. This challenge could also

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serve as a motivation for the researchers to develop their own printers with desired functionalities. As an example, our lab adapted commercially available components to build a custom printer for liquid metal.[59]

Finally, we identified future trends and technologies that could drive the innovation in this field in the recent future. We predict that a major area of focus will now be printing multilayered structures with different materials to create a richer set of functionality. This should give rise to a system-based approach toward printing in which ICs and other components get integrated (or printed) with the stretchable electronic components for on board computational power. This approach should also drive innovations in printed stretchable power sources such as super capacitors and batteries. We envisage that development of news material and printing strategies will form the bedrock of these developments. For instance, integration with rigid ICs would benefit from printing of stretchable interconnects with variable stiffness to create gradients in mechanical properties (and thereby avoid sharp interfaces between soft and hard materials). Further, in multifunctional devices, these interconnects will also have to be thermally stable to be able to sustain high current density especially for high resolution printing. On the printing side, promising methods such as embedded printing and meniscus guided printing are being studied for printing stretchable conductors. For printing stretchable conductors on nonplanar substrates, new technologies such as robotic arm integrated direct writing system offer higher degree of freedom in printing supported and free-form structures. In this pursuit of innovations in the field of printed stretchable conductors, we hope the structured methodology presented in this guide will benefit researchers.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.chemrev.3c00569

References to the entries in Ashby plots in Figure 2 (PDF)

AUTHOR INFORMATION

Corresponding Author

Michael D. Dickey − *Department of Chemical and Biomolecular Engineering, North Carolina State University, Raleigh, North Carolina 27695, United States;* orcid.org/0000-0003-1251-1871; Email: mddickey@ncsu.edu

Authors

Tushar Sakorikar − *Department of Chemical and Biomolecular Engineering, North Carolina State University, Raleigh, North Carolina 27695, United States*

Nikolas Mihaliak − *Department of Chemical and Biomolecular Engineering, North Carolina State University, Raleigh, North Carolina 27695, United States*

Febby Krisnadi − *Department of Chemical and Biomolecular Engineering, North Carolina State University, Raleigh, North Carolina 27695, United States*

Jinwoo Ma − *Department of Chemical and Biomolecular Engineering, North Carolina State University, Raleigh, North Carolina 27695, United States;* orcid.org/0000-0001-5140-2972

Tae-il Kim − *Department of Chemical and Biomolecular Engineering, North Carolina State University, Raleigh, North Carolina 27695, United States; School of Chemical Engineering, Sungkyunkwan University (SKKU), Suwon, Gyeonggi 16419, South Korea;* orcid.org/0000-0002-6667-5356

Minsik Kong − *Department of Chemical and Biomolecular Engineering, North Carolina State University, Raleigh, North Carolina 27695, United States; Department of Materials Science and Engineering, Pohang University of Science and Technology (POSTECH), Pohang 37673, Republic of Korea*

Omar Awartani − *Department of Chemical and Biomolecular Engineering, North Carolina State University, Raleigh, North Carolina 27695, United States*

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.chemrev.3c00569

Author Contributions

CRediT: **Tushar Sakorikar** data curation, formal analysis, visualization, writing-original draft, writing-review & editing; **Nikolas Mihaliak** data curation, methodology; **Febby Krisnadi** investigation, visualization, writing-review & editing; **Jinwoo Ma** data curation, formal analysis, writing-review & editing, **Tae-il Kim** formal analysis, writing-review & editing, **Minsik Kong** writing-review & editing, formal analysis; **Omar Awartani** conceptualization, data curation, visualization, writing-review & editing; **Michael D. Dickey** conceptualization, funding acquisition, project administration, writing-review & editing.

Notes

The authors declare no competing financial interest.

Biographies

Tushar Sakorikar received a B.E. in Electronics and Communication Engineering from RGPV, India (2010) and an MS-PhD from the Indian Institute of Technology Madras (2019) under the guidance of Prof. Manu Jaiswal and Dr. Pramitha V. From 2020-2022, he was a SERB National Postdoctoral fellow at Indian Institute of Science, Bangalore in the lab of Prof. Hardik J. Pandya. Since 2022, he has been working as a Fulbright-Nehru Postdoctoral Fellow in the lab of Prof. Michael Dickey at North Carolina State University. His research interests include the study of soft materials for stretchable electronics and bioelectronic devices.

Nikolas Mihaliak received his B.S. in Computer Engineering from North Carolina State University in 2021 and went on to pursue a Master of Engineering Management degree from Duke University in 2023. He now works at Applied Research Associates. His research interests include composite research used in stretchable electronics and nanofabrication techniques applied to electrical systems.

Febby Krisnadi received a B.Eng. in materials engineering from Nanyang Technological University (2020) under the guidance of Professor Nripan Mathews. She is currently a Ph.D. student advised by Professor Michael Dickey at NC State University. Her research interests include multiphasic composites, functional pastes, and electromechanical properties of soft materials.

Jinwoo Ma received a B.S. degree in materials science and engineering from Seoul National University, Seoul, South Korea, in 2012, and a Ph.D. degree in materials science and engineering from Seoul National University, in 2020, under the supervision of Prof. Kyu Hwan Oh and Jeong-Yun Sun. He has been a visiting scholar at the Dickey Group, North Carolina State University, Raleigh, NC, USA, from 2017 to 2021 and currently works in Samsung Electronics. His research interests include patterning and actuating liquid metals for flexible and stretchable electronics and actuating soft materials by harnessing smart polymer and interfacial bonding.

Tae-il Kim received a BS in Chemical Engineering from Sungkyunkwan University (SKKU) (2003) and a PhD from the Seoul National University (2009) under the guidance of Professor Hong H. Lee. From 2009-2012 he was a postdoctoral fellow in the lab of Professor John A. Rogers at University of Illinois at Urbana−Champaign. He is currently Professor in the School of Chemical Engineering at SKKU (Korea). His research are bioelectronics and biomimetics with soft materials and unconventional patterning techniques.

Minsik Kong received a B.S. in materials science and engineering from Pohang University of Science and Technology (POSTECH) in 2016 and a Ph.D. from the same department under the supervision of Prof. Unyong Jeong. He is now a postdoctoral researcher in the same group. His research interests include interface issues in soft electronic devices and the characterization of amorphous oxides formed over liquid metals.

Omar Awartani received his B.Sc. in Aerospace Engineering from NC State University. In 2010, he joined Prof. Brendan O'Connor's group at NC State University, where he completed his M.Sc. and Ph.D. in Mechanical Engineering. From 2015-2017, he was a postdoctoral fellow in Prof. Harald Ade's group. In 2019, he joined the American University of Beirut as an assistant professor in mechanical engineering and was a visiting professor at Prof. Michael Dickey's lab from 2020-2022. Currently, he is a research scientist and technical lead at Meta Reality Laboratories-Research. His research interests include flexible and stretchable electronics, patterning of liquid metals, conductive inks, and flexible/stretchable integrated electronics.

Michael Dickey received a BS in Chemical Engineering from Georgia Institute of Technology (1999) and a PhD from the University of Texas (2006) under the guidance of Professor Grant Willson. From 2006-2008 he was a postdoctoral fellow in the lab of Professor George Whitesides at Harvard University. He is currently the Camille and Henry Dreyfus Professor in the Department of Chemical & Biomolecular Engineering at NC State University. He completed a sabbatical at Microsoft in 2016. Michael's research interests include soft matter (liquid metals, gels, polymers) for soft and stretchable devices (electronics, energy harvesters, textiles, and soft robotics).

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ADDITIONAL NOTES

[1] In Figure 2a, the roll printed conductors are flexible and not stretchable. These values of viscosities are plotted to provide a range of values for the reader. Further, for liquid metal although these are 'bulk' viscosity values, it is important to note that the rheology of liquid metal is more complex due to the presence of the oxide.

[2] The two references used in Figure 2b for roll-printing have not reported viscosity values. Hence, they were excluded from the comparison in Figure 2a.

[3] Stencil printing has been suggested to have historically evolved from screen printing.[102] Therefore, stencil printing can be treated as a subset of screen printing.

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