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Topical Review

A review of non-contact micro- and nano-printing technologies

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Abstract

Printing technologies have undergone signficant development because they are an enabler in science and engineering research; they also have significant practical applications in manufacturing. Micro- and nano-printing techniques have found a number of applications in electronics, biotechnology, and material synthesis/patterning. In this review, we look at the important printing methods, including high precision traditional printing methods as well as recently emerging techniques. We also discuss the materials that are printable by these technologies, the challenges for future development, and the applications of micro- and nano-printing.

Keywords: nano-printing, micro-printing, 3D printing

(Some figures may appear in colour only in the online journal)

1. Introduction

A variety of industries utilize printing-based manufacturing. Traditionally, printing has been used for the production of newspapers and books. A number of new printing methods have emerged recently, and many new printing materials have been developed. As printing technologies progress, printing precision can reach micrometer and sub-micrometer scales. These new technologies enable such applications as displays, biochips, drug delivery, and functional material deposition.

The development of micro and nano-scaled printing technologies is partly motivated by the unique properties of materials as they miniaturize. These materials have significantly different properties at small scales compared to the bulk state. For instance, gold nanoparticles have a lower melting point than bulk gold, enabling the use of printing to deposit gold nanoparticles on microelectronic circuits that can then be sintered onto the circuits at a relatively low temperature. The printing of metal or semiconductor nanoparticles is particularly relevant for the manufacturing of flexible electronics. New printing technologies have also advanced many aspects of biotechnology, particularly in biosensors and microarrays.

Traditional printing methods include thermal and piezoelectric printing, as well as the electrostatic printing. However, these traditional methods are limited in precision. With the advent of electrohydrodynamic jet printing, droplets that are smaller than the nozzle itself can be printed at room temperature and at a high speed. To reduce the complexity in electrohydrodynamic jet printing, surface acoustic wave (SAW) based printing and pyroelectrodynamic printing have been developed, eliminating the need for a jet nozzle. This review will provide comprehensive discussions of these and

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many other printing methods, materials for printing, and their applications.

2. Printing-based manufacturing methods

2.1. Inkjet printing

The widely used methods in the ink-jet market are thermal and piezoelectric inkjet printing. Both have had significant commercial success in digital printing. Early inkjet printers used binarily operated electrostatic fields to control ink drop dispensing [1]. Kyser and Sears [2] invented piezoelectric inkjet printing, and Endo and Hara [3] invented DOD ink-jet printing (i.e., the bubble jet), which later evolved into thermal inkjet printing.

2.1.1. Piezoelectric inkjet printing. Piezoelectric inkjet printing is based on the deformation of many tiny piezoelectric ceramics. Voltages applied to the ceramics induce mechanical vibration to overcome the liquid surface tension for droplet ejection. Creagh and Frost [4] reviewed recent advances in the piezoelectric drop-on-demand (DOD) inkjet technology, especially for application in flat panel displays and organic electronic devices. Percin and Khuri-Yakub [5] demonstrated a technique for the printing of fluids and solid particles using a piezoelectric droplet ejector by bonding a thin piezoelectric annular disc to a thin edge clamped circular plate. The size of the droplets ejected ranged from 5 μm at 3.5 MHz to 150 μm at 7 kHz [6, 7]. Unlike thermal inkjet printing, piezoelectric ejectors are compatible with thermally sensitive solutions.

2.1.2. Thermal inkjet printing. Thermal inkjet printing is based on tiny resistors in a chamber that is filled with ink. These resistors act as heating elements. When a voltage is applied, the heating elements cause a sudden steam explosion in the ink chamber, which produces vapour bubbles. As the bubbles expand, they push the droplets of ink out of the nozzle and onto a substrate. Chen and Wise [8] demonstrated a high-resolution monolithic nozzle array for thermal inkjet printing. The nozzle array minimizes heat dissipation to the substrate and enhances the energy efficiency for ink actuation.

Thermal and piezoelectric inkjet printing both rely on actuators to push droplets through a nozzle. A thermal bubble actuator generates heat, which can cause print head damage at high temperatures for prolonged printing periods and is unable to operate at high frequencies. Thermal bubble actuators and piezoelectric actuators are unable to dispense droplets smaller than the nozzle size. The head, consisting of piezoelectric or heater elements, also has a complex structure [9]. The printing resolution of thermal or piezoelectric printing is also typically limited to micrometers.

2.1.3. Electrostatic inkjet printing. Electrostatic inkjet printing is based on a change of the ink volume due to the deformation of the chamber. The chamber has two apertures, one for droplet ejection and the other for ink to enter the chamber. Ink enters the chamber due to chamber deformation, which is caused by the application of voltages. When the voltage is removed, the chamber returns to the original shape, causing the ink droplet to be ejected. Kamisuki *et al* [10, 11] developed a static-electricity actuator inkjet (SEA-JET), which is a thin and small inkjet head driven by electrostatic force. The head was fabricated from batch micromachining and a parallel-plate actuator mechanism, which can be scalable to operate at a high frequency with a high density of nozzles. Compared to conventional thermal and piezoelectric inkjet printing, the SEA-JET has the advantages of lower power consumption, it is smaller in size, and it has a higher mass productivity. Lee *et al* [12] reported the use of electrostatic forces to print droplets of a charged nanoparticle colloid.

2.1.4. 3D inkjet printing. Three dimensional (3D) printing allows the making of objects of virtually any shape from a design model. It is an additive process involving the printing/deposition of successive layers of material in different shapes. In 2D inkjet printing, since it is mostly used for graphics the height of the deposited material is less important than the width of the printed pattern. In contrast, in 3D inkjet printing accurate control is needed in three orthogonal directions. The main difference between 2D and 3D inkjet printing is that in 3D printing the print head or the surface being printed is controlled to move both vertically and horizontally, and it dispenses waxes and polymers to construct an object layer by layer.

Traditional photolithography/etching and 2D ink writing/printing are cost intensive and confined to two dimensions [13]. 3D inkjet printing of functional materials enables low-cost, large-area device construction with high throughput [14]. Schiaffino and Sonin [15] numerically and experimentally studied the 3D inkjet printing of micro-droplets of different fluids, including wax, water and mercury. Haferl and Poulikakos [16] demonstrated that the pileup of molten micro-droplets can be used to fabricate 3D micro-scale structures by inkjet printing. Since 3D inkjet printing requires low temperature, it is also suitable for fabricating electronics on plastic substrates. Yamaguchi [17] first demonstrated the 3D metal jetting of molten solder. However, the molten metal jet process is limited because it uses low-melting-temperature solder and needs high-temperature resistant systems to keep the metal molten during the jetting process [18]. A low-temperature 3D micro metal structure fabrication by direct inkjet printing of metal nanoparticles has also been reported [19].

2.2. Electrohydrodynamic printing

Electrohydrodynamic printing (e-jet printing) employs an electric field-induced flow through microcapillary nozzles to deliver the ink onto a substrate. Figure 1 is a schematic diagram of a typical e-jet printing system. A syringe pump is connected to a glass capillary to deliver fluidic ink. A voltage is applied between the nozzle and substrate to drive the ink onto the target substrate. The nozzle, having an inner diameter of a few micrometers, can be fabricated by a standard micropipette puller. The nozzle is then metal coated to make it electrically conductive. The substrate must be a metallic (e.g.,

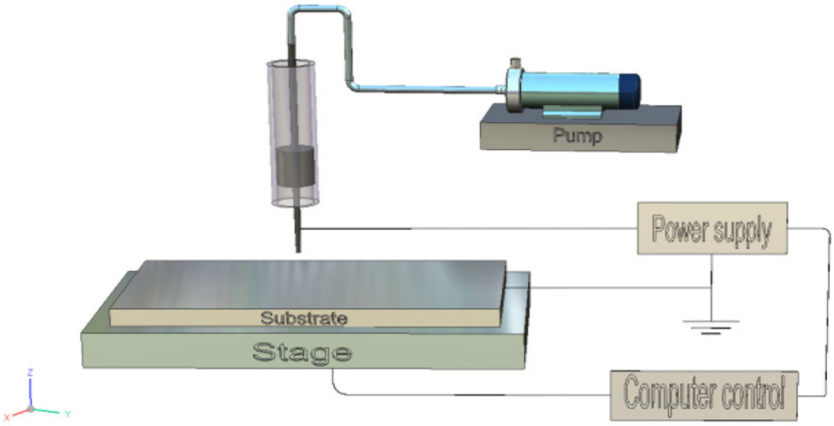


Figure 1. Schematic diagram of a typical e-jet printing system consisting of a metal-coated nozzle, an ink chamber, a pump, a conductive substrate, a translational stage, and a power supply.

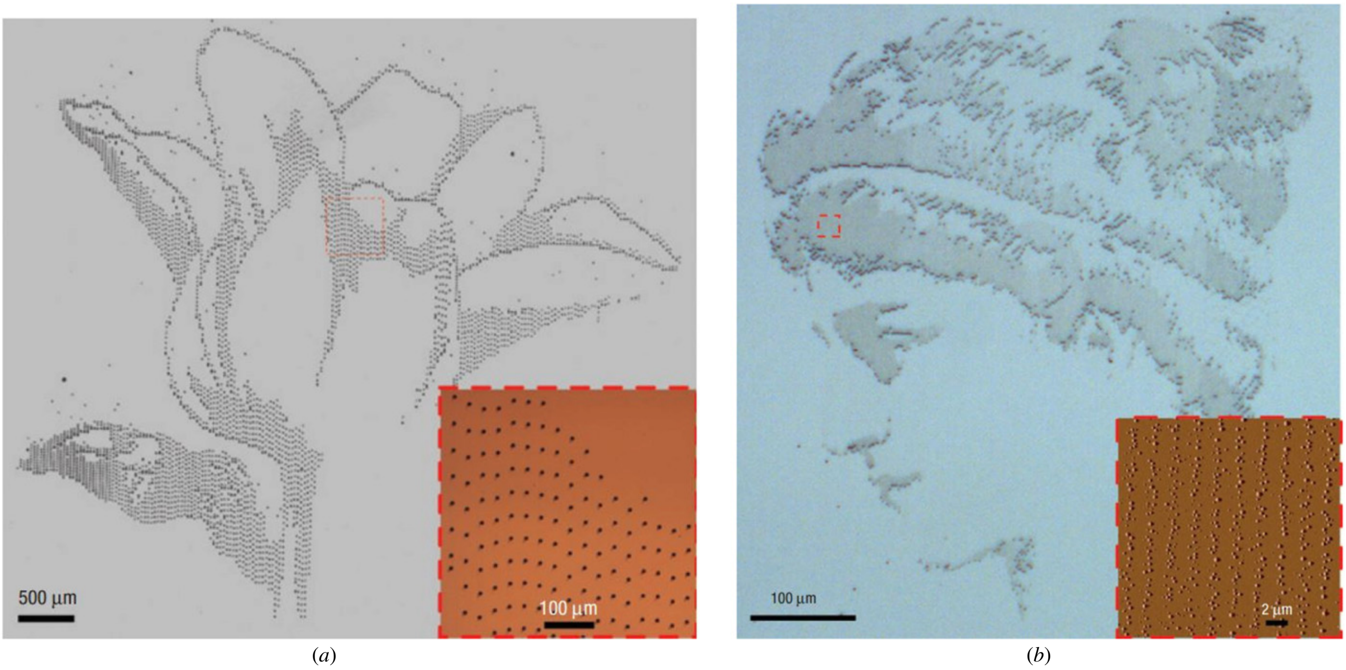


Figure 2. (*a*) Image of a flower printed by e-jet printing. The dots (about 10 μm in diameter) are SWNTs, printed with a nozzle with internal diameter of 30 μm. (*b*) A portrait printed using polyurethane ink and a nozzle with 500 nm internal diameter. The individual dots are about 500 nm in size. (Reprinted from Park *et al* [24] with permission).

a copper sheet) or semiconductive (e.g., a Si wafer). The dc power supply outputs an adjustable high voltage. When an appropriate voltage is applied between the metal-coated glass tip and the conductive substrate, an electric field is generated and this drives the ions to the nozzle. As the ions in the ink accumulate near the tip of the nozzle, mutual Coulomb repulsion between the ions causes a tangential stress on the liquid surface, thereby deforming the meniscus into a conical shape, which is known as the Taylor cone [20, 21]. Electrostatic stress overcomes the surface tension at the tip of the cone, causing the ejection of the ink from the tip of the already existent cone. By varying the applied voltage, the pump's pressure, the stand-off height, and the fluid properties, the setup can generate different jetting modes, such as pulsating, stable jet, and e-spray mode for various printing or spraying applications [22, 23].

2.2.1. High-resolution electrohydrodynamic jet printing. Park *et al* [24–26] first adapted the electrohydrodynamic method to print high-resolution patterns. Figure 2(*a*) shows an image of a flower that has been made of printed dots of single-walled carbon nanotubes (SWNTs) [27] in an aqueous solution by using e-jet printing. These dots have an average diameter of approximately 10 μm, and they are ejected from a nozzle with an internal diameter of 30 μm. A fine internal diameter is important for achieving high resolution. Park *et al* also used a much finer nozzle (500 nm inner diameter) to generate dots of about 490 nm (figure 2(*b*)).

Although high-resolution electrohydrodynamic jet printing overcomes the resolution limitation of conventional ink-jet printing [28], the construction of micrometer and sub-micrometer-sized metal-coated nozzles with a high consistency involves a certain complexity and intricacy.

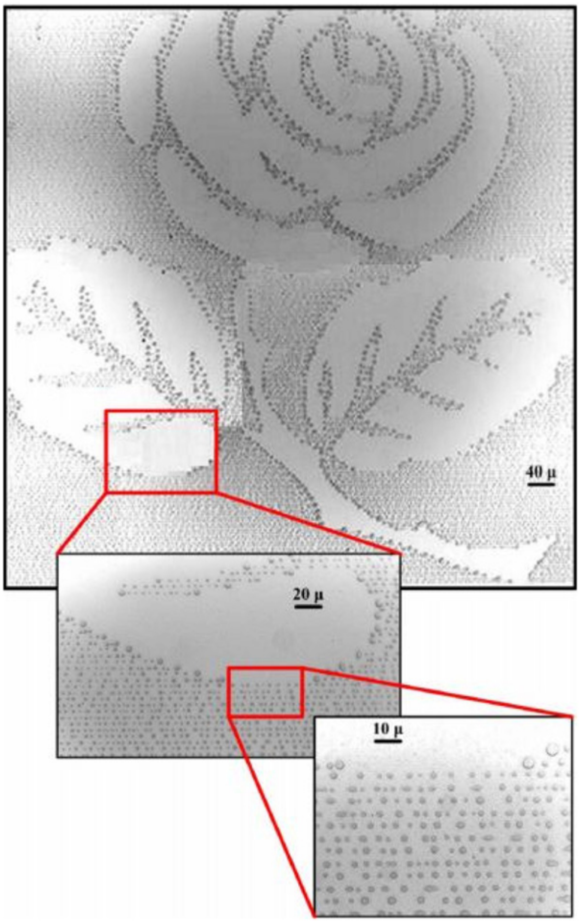


Figure 3. Flower image with dot diameters of 1–2 μm, printed using photocurable polyurethane polymer at 1 kHz using a nozzle of 2 μm internal diameter. (Reprinted from Mishra *et al* [22] with permission).

In e-jet printing, different voltages can cause significant variations in jetting frequency and droplet size. While too low a voltage results in lower printing speeds, the application of too high a voltage induces larger variations in droplet uniformity and spacing.

2.2.2. Pulsed e-jet for high speed and drop-on-demand printing. Instead of applying a constant voltage, Mishra *et al* [22] used pulsed dc voltage for e-jet printing to realize DOD, high speed, high resolution printing. Pulse duration determines the volume and number of droplets. The period between pulses determines the distance between droplets. In between pulses, the positioning stage moves at a constant velocity. By choosing a suitable pulse width and frequency, high-speed (1 kHz) DOD printing with a droplet size within 3–5 μm for an aqueous ink and 1–2 μm for a photocurable polymer ink was demonstrated [22]. Figure 3 shows an image printed at 1 kHz with droplet sizes ranging from 1–2 μm. Kim *et al* [29, 30] used squarewave dc pulses for e-jet printing and used piezoelectric excitation to jet. Stachewicz *et al* [31, 32] reported single-event pulsed droplet generation for a DOD e-jet. Pulsed e-jet printing technology has shown its potential for application in microelectromechanical systems and biotechnology due to its high printing speed, DOD capability, precise droplet size control, and precise droplet placement.

2.3. Pyroelectrodynamic printing

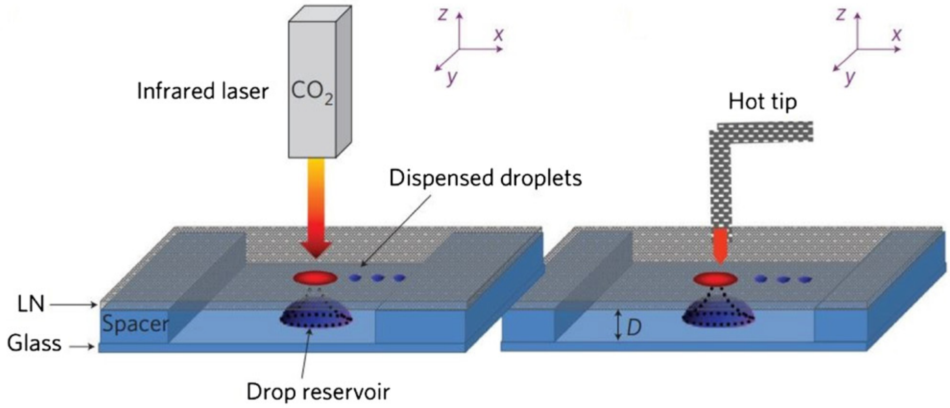
When a liquid is subject to a sufficiently strong electric field, it can be induced to emit thin fluid jets [33]. Instead of e-jet printing using a fine printing nozzle and high voltages, Ferraro *et al* [34–39] demonstrated a technique that draws attolitre liquid droplets from one drop or liquid film reservoirs via the pyroelectric effect. This system does not require electrodes or nozzles, as shown in figure 4. The pyroelectrodynamic printing system consists of two plates and a heat source. The heat source can either be an infrared laser beam or a hot tip. The infrared laser beam provides more flexibility since laser-generated heat can be better controlled, and the laser beam can be split to achieve simultaneous multiple droplet dispensing. In figure 4, a microscope glass slide was used as a liquid reservoir and the lithium niobate (LN) crystal wafer was used as the target substrate. The droplets dispensed in their system have an average radii of about 300 nm. In contrast to conventional methods and e-jet printing [40, 41], which require fine nozzles, this pyroelectrodynamic printing is nozzle-free.

In the pyroelectrodynamic printing system, when a pointwise thermal stimulus is applied to the substrate, the heating initiated by the thermal stimulus induces a polarization that brings about uncompensated charges on the surface of the substrate. When the LN substrate is placed over another substrate to support ink droplets or films of liquid, the pyroelectric effect in the LN substrate can generate the electrohydrodynamic phenomenon. When the electric fields exceed a critical value, pulsating jets begin similarly to e-jet printing in that there are forces pulling the liquid from the substrate [42]. The liquid tip is similar to the Taylor cone [20, 24, 43], and it should be pointed out that the liquid is not conductive.

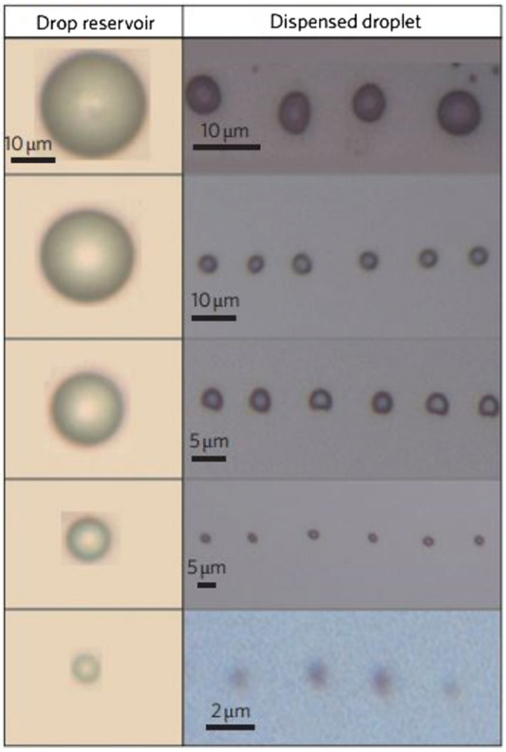
Ferraro demonstrated that this technique is able to print droplets with smaller sizes by decreasing the volume of the drop reservoir. Figures 4(*b*) and (*c*) show the approximate relationship between the volume of the drop reservoir and the radii of the printed droplets. The smallest droplets printed by this system reached about 300 nm. With this method, many materials (such as metal particles in oil) can also be printed. Eliminating the need for an actual nozzle, electrodes or high-voltage power, enables this technique to avoid nozzle-induced cross contamination. This, together with its high-speed, makes this technique particularly attractive in the fabrication of assays for use in chemistry and biology [44–47], and in the construction of printed electronics.

2.4. Dielectrophoretic printing

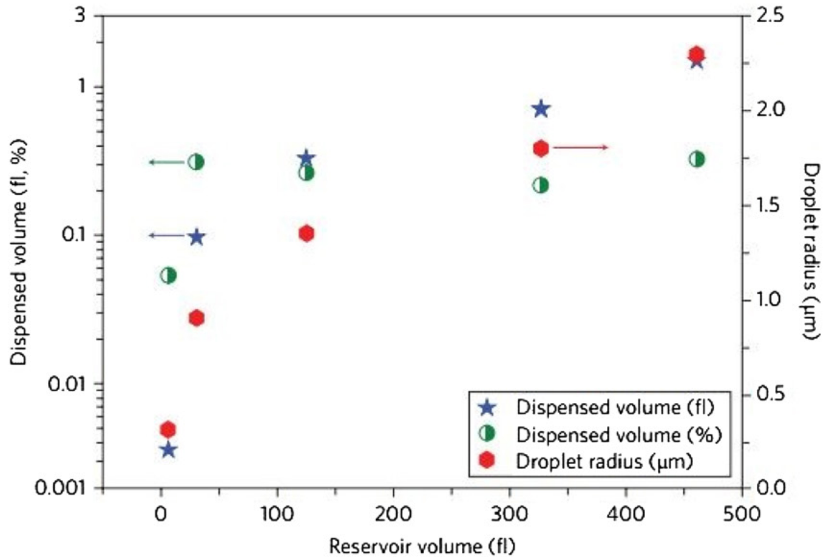
Schirmer *et al* [48] demonstrated on-demand dielectrophoretic nanoprinting, based on the effect of dielectrophoresis (DEP) [49]. DEP has been used for nanotube manipulation and particle alignment [50, 51]. In DEP printing, dielectrophoretic forces are generated on the colloidal particles suspended in the dispensing liquid. Schirmer *et al* deposited gold nanoparticles to evaluate the precision of the printing process. As shown in



(*a*)



(*b*)



(*c*)

Figure 4. (*a*) Schematic of the pyroelectrodynamic printing system, including two plates and a heat source. (*b*) Images of different drop reservoirs and printed droplets. (*c*) Relation between the volume of the drop reservoir and the radii of droplets. (Reprinted from Ferraro *et al* [34] with permission).

figure 5, a dc voltage is applied between the substrate and the tip of a metal-coated nozzle. A capillary with a small orifice diameter is filled with a liquid that has gold nanoparticles. A sufficiently large DEP force can result in the ejection of the colloidal liquid. The nanopaticles are driven towards the tip of the nozzle. The pulsation of the dc field is the same as in electrohydrodynamic printing [24] and this enables the controlled printing of nanoparticles [48]. By using a 150 nm head nozzle and proper pulse lengths, dots of 175 nm in diameter were deposited on a silicon substrate. Ishida *et al* [52] demonstrated the ejection of droplets from a silicon needle inkjet head using an electrostatic inkjet setup. Silicon needles fabricated with the anodization technique have a high aspect ratio and sharp tip, and can produce sub-micrometer patterns.

2.5. SAW Printing

SAW has been used in RF communication, optical modulation, and biochemical sensors. It is also effective for the generation of micron and sub-micron droplets. SAW manipulates droplets without the use of nozzles and orifices, a characteristic which is similar to pyroelectrodynamic printing. The advantages of SAW printing are that it uses large forces, and fast and simple electrode networks. SAW uses acoustic fields to drive the fluid's movement. These waves are nanometer order amplitude

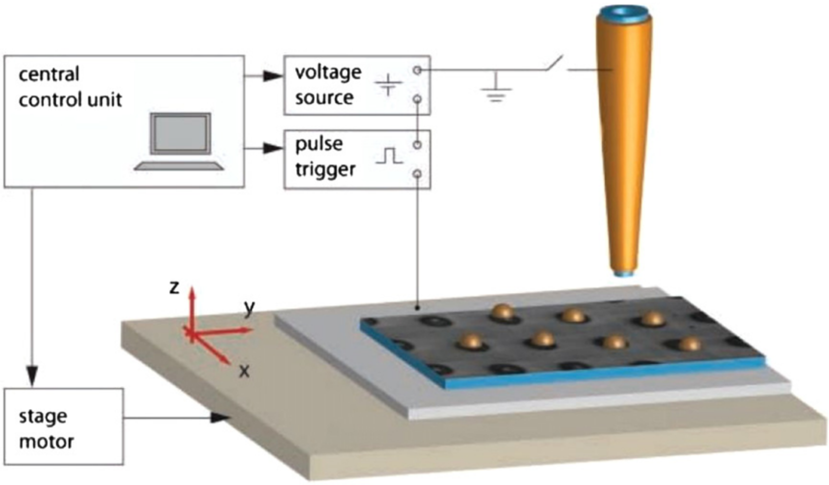


Figure 5. Schematic of a dielectrophoretic (DEP) printing system consisting of a metal-coated microcapillary nozzle, a substrate, a motion stage, a dc power supply, and a controller. (Reprinted from Schirmer *et al* [48] with permission.).

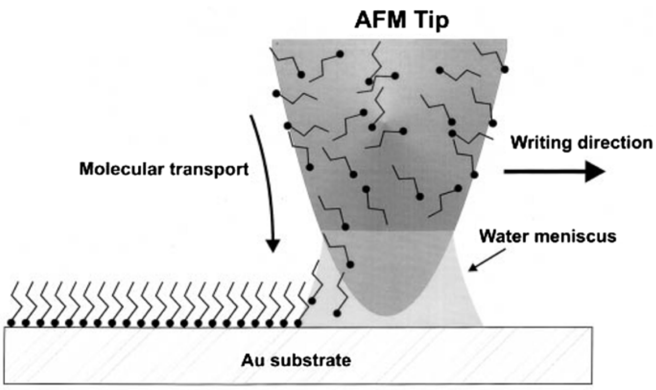


Figure 6. Dip-pen nanolithography (DPN) writing. (Reprinted from Piner *et al* [57] with permission.)

electroelastic waves that spread along the surface of an elastic material's substrate. Acoustic waves can generate pressure gradients and acoustic forces in a liquid for dispensing/jetting.

Tan *et al* [53] reported the generation of two series of identical SAWs propagating in opposite directions at a point where a droplet is located. The capillary waves at the droplet surface overcome capillary stress and result in atomization of the droplet when the applied power is sufficiently high. However, no printing result was demonstrated, and the ink drop size was 100–200 μm in diameter. This size is larger than the state-of-the-art ink jet printing. The best high resolution pattern SAW was demonstrated in the printing of 100 nm protein nanoparticles on a substrate with SAW atomization [54–56]. However, this approach is unable to create free form geometries, such as text or pictures.

2.6. Pen printing/desposition

2.6.1. Dip-pen nanolithography. Dip-pen technology utilizes capillary forces to carry ink on a sharp tip to a substrate. Dip-pen nanolithography (DPN) was first demonstrated by Piner *et al* [57]. As shown in figure 6, the tip of an atomic force microscope (AFM) probe is used as a nib. Molecules are printed to the Au substrate with nanometric dimensions via capillary forces during the microcontact between the substrate and probe tip. Piner *et al* [58] studied the parameters that affected the resolution of DPN printing, finding that grain sizes of the substrate, choice of ink, tip-substrate contact time, scan speed, and relative humidity all contributed to the resolution. DPN has been used for structure creation on the nano/sub-microscale to create biological architectures. For example, Thompson *et al* [59] used DPN to push analytical limits in proteomic concentration detection. Electrochemical DPN (E-DPN) is an extension of DPN. By applying a voltage between the tip of the AFM probe and the conducting substrate, metal salts were dissolved in the water meniscus and deposited on the substrate [60]. DPN has become a useful approach for the nanofabrication of a wide variety of metal [61–63] and semiconductor [64, 65] structures, as well as for the deposition (printing) of organic molecules [66–69], DNA [70–72], polymers [73, 74] and proteins [61, 72].

2.6.2. Nano fountain pen. The nano fountain pen (NFP) technique uses a glass or quartz capillary that has a tapered tip and an aperture of a few hundred nanometers to deliver liquid from the capillary onto a target substrate. When liquid fills the capillary, it flows to the end of the tapered tip by capillary forces. The liquid flows out of the capillary only when it contacts the substrate due to the surface tension of the droplet formed at the end of the capillary. Taha *et al* [75] used a tapered micropipette tip to deposit nanometric dots (∼300 nm diameter) on a gold-coated substrate by employing near-field optical microscopy. This technique can also be useful in printing DNA patterns with a high resolution. Choi *et al* [76] demonstrated the use of NFP to deposit ink with gold nanoparticles (2–4 nm). Belmont *et al* [77] wrote structures of molecularly imprinted polymers with the thickness of dots in the range of tens of nanometers for the fabrication of highly integrated biomimetic nanochips. Onoue *et al* [78] demonstrated the fabrication of photomasks by fountain-pen nanolithography using ink filled with silver nanoparticles. Compared to DPN, fountain-pen nanolithography is capable

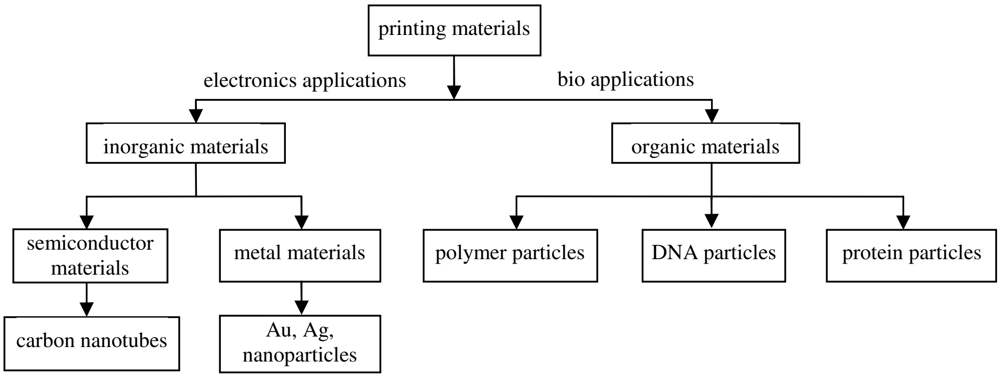


Figure 7. Example materials that can be printed with advances in printing technologies.

of continuous printing without the need for repeated inking since an ink reservoir is connected to the capillary tube in the NFP for refilling.

3. Printing materials

With the advances of printing technologies, the materials that can be printed have become more diverse, for instance: metal particles, polymer particles, proteins, DNA, nanotubes, and nanowires. We have classified examples of the printable materials according to their applications, as shown in figure 7.

3.1. DNA and proteins

The DNA microarray has become an indispensable tool in genomics and medical diagnostics. Thermal and piezoelectric inkjet printing has been used for DNA microarray construction due to their noncontacting nature and their compatibility with DNA [79]. Typical resolutions of thermal and piezoelectric inkjet DNA printing is about 10 μm. Park *et al* [24] reported the use of high-resolution electrohydrodynamic jet printing to form dots of double-stranded DNA with diameters less than 100 nm in the microarrays. DPN using an AFM probe tip has also formed DNA patterns, with DNA droplet sizes smaller than 50 nm [80, 81]. Similar to DNA printing, the printing of proteins is also critical for the fabrication of biochips. Taha *et al* [75] demonstrated that protein dots (200 nm) can be printed through a nanopipette having an aperture of 100 nm, via NFP printing.

3.2. Metal nanoparticles

For applications in electronics and phonotics, nanoparticles (e.g., Au and Ag) are important functional materials due to their high conductivity, chemical stability, and resistance to surface oxidation [82–85]. Samarasinghe [86] used electrohydrodynamic printing to deposit Au nanoparticles in the liquid phase onto a substrate. The average size of printed droplets was $\sim$26 μm. Sekitani *et al* [87] manufactured organic transistors with sub-femtoliter accuracy using inkjet technology, and overcame the obstacles between manufacturing efficiency, transistor performance, and power consumption by creating high-performance transistors of p-channel and n-channel and low-power complementary circuits. Khan *et al* [88] demonstrated the e-jet printing of a Ag colloidal solution for the fabrication of electrically functional microstructures. Using multiple nozzles in their electrohydrodynamic inkjet printing, they increased printing throughput.

3.3. Polymer particles

Polymer nanofibres have been shown to have unique characteristics for applications, such as chemical sensors [89] and drug delivery [90]. Nanofiber biomaterials fabricated through printing have attracted much attention and shown great promise in skin, vascular, and neural tissue engineering applications. Sun *et al* [91] printed polyethylene oxide nanofibres (100 nm to 800 nm) on a silicon substrate based on electrospinning and investigated the behaviour of the nanofibres. Park *et al* [24] used nanoscale nozzles to print complex patterns of positive and negative charges (the ink consisted of photocurable polyurethane) using electrohydrodynamic printing. The resolution of the patterns reached the nanometer regime. The use of a printed charge provides an application example of the control of the properties of nanoscale electronic devices. Sokuler *et al* [92] presented the printing of polymer micro-lenses via NFP to yield microlenses with diameters in the range of 4–9 μm. It was demonstrated that the microlenses can be useful on integrated biochips due to their high transparency, low weight, and cost [93–95].

4. Applications

This review has discussed recent advances in micro-nanometer scaled printing technologies. The major characteristics of each printing technology are summarized in table 1, and their advantages and disadvantages are summarized in table 2. These printing technologies overlap in certain capabilities but differ in many aspects, such as resolution, simplicity in system setup, capability for scaled-up operation, and materials suitable for printing.

Thermal and piezoelectric printing [96, 97] has a longer history and is typically limited to a resolution of micrometers. Wang *et al* [98] demonstrated a high-resolution monolithic thermal bubble inkjet print head and achieved sub-micrometer printing resolution. The inkjet printing and other printing techniques discussed in this review form a repertoire of

Table 1. Characteristics of each printing method.

Printing methods		Precision	Equipment	Need nozzle?	Power consumption	Contact or non-contact	Printable materials
Inkjet methods	Thermal inkjet	Micron	Complex	Yes	High	Non-contact	Polymers, metal materials
	Piezoelectric inkjet	Micron	Complex	Yes	High	Non-contact	Polymers, metal materials
	Electrostatic inkjet	Submicron	Complex	Yes	Low	Non-contact	Polymers, metal materials
	3D inkjet printing	Micron	Complex	Yes	High	Non-contact	Polymers, ceramic, glass materials
Electrohydrodynamic printing		Submicron	Simple	Yes	Low	Non-contact	Polymers, DNA, proteins, metal materials
Pyroelectrodynamic printing		Submicron, nano	Simple	No	Low	Non-contact	Nanotubes/nanowires, mineral oil
Dielectrophoretic printing		Submicron, nano	Simple	Yes	Low	Non-contact	Polymers, DNA, proteins, metal materials
SAW printing		Micron, submicron, nano	Simple	No	Low	Non-contact	Polymers, metal materials
Dip-pen nanolithography		Submicron, nano	Simple	No	Low	Contact	Polymers, DNA, proteins, metal materials
Nano fountain pen		Submicron, nano	Simple	No	Low	Contact	Polymers, DNA, proteins, metal materials

Table 2. Advantages and disadvantages of each printing method.

Printing methods	Advantages	Disadvantages
Thermal inkjet printing	On-demand printing widely used in practical applications	Complex structure [93] difficult in the sub-micrometer range typical micrometer resolution easy to damage sensitive solution [91] short working life
Piezoelectric inkjet printing	On-demand printing widely used in practical application ejector does not damage sensitive solution [91]	Complex structure [93] difficult in the sub-micrometer range typical micrometer micrometer resolution short working life
Electrostatic inkjet printing	High mass productivity [94], low power consumption eject fine droplets [100]	Unable to fine control droplet size short working life
Electrohydrodynamic printing	High-resolution, simple setup, precise droplet placement [24]	Intricate to precisely control droplets at the tip of the Taylor-cone [57] charge deposition on surface complex fabrication of nozzle prone to cross-contamination [45]
Pyroelectrodynamic printing	No need for an actual nozzle and electrodes high-voltage power avoid the cross-contamination high-speed printing potential [45]	Early stage development hot tip tends to damage droplet reservoir exposed to atmosphere
Dielectrophoretic printing	Control precisely on-demand printing of nanoparticles ultrahigh resolution [54]	Early stage development complex fabrication procedures of gold-coated nozzle
Printing based on surface acoustic wave	Without the nozzles and orifices reduce equipment cost simple structure [59, 70]	Developed for automatic manipulate in practical application
Dip-pen nanolithography	Submicron dimensions of printed droplets precision control directly write nanowires [71]	Many factors affect resolution difficult to control [71, 72] high equipment requirements low throughput
Nano fountain pen	Submicron resolution print solution of many types of molecules [81, 82]	High equipment requirement high cost low throughput

technologies that have enabled a multitude of applications. For instance, printed electronics, also known as flexible electronics or organic electronics, involves the production of electronic devices of organic or inorganic materials via printing [99–101].

The inkjet printing of functional materials enables the fabrication of low-cost, large-area printed electronics. Selective laser sintering and ablation of inkjet-printed metal nanoparticles enables low-temperature metal deposition, as well as high resolution patterning [102]. High-resolution organic transistors have been fabricated in ambient pressure and room temperature without utilizing photolithographic steps or requiring a vacuum deposition process [103]. Inkjet printing can also deposit seed nanoparticles for selective nanowire growth, which can be applied to flexible plastic substrates and scaled up for larger substrates for mass production or roll-to-roll processing. The selective growth of ZnO nanowire arrays has been achieved from inkjet printed zinc acetate precursor on a heated substrate [104, 105]. Other device examples are thin film solar cells [87, 88, 106, 107] and thin film transistorsTFT [108, 109]. Fujihara *et al* [110] fabricated TiO_2 nanofiber electrodes for solar cells by using electrospraying [111].

As opposed to printing on paper or textile, which absorbs the ink, material deposition on glass or polymer substrates requires solvent evaporation. Therefore, heating the substrate can quicken solvent evaporation, and a thicker layer can be deposited. Rapid liquid evaporation also eliminates uneven wetting problems and achieves smaller feature sizes [112]. However, inkjet printing on a heated substrate causes the temperature to rise in the nozzle due to conductive heat diffusion from the hot substrate through air. Therefore, it is beneficial to reduce temperature rise in the nozzle by integrating an active cooling system into the jetting head.

3D inkjet printing has found widespread use in electronics and bioengineering. Precise 3D inkjet printing of conductive copper has been achieved with low cost and low material waste for circuit board production [113]. Lithium ion batteries have been 3D printed for energy storage [114]. 3D inkjet printing has also been used to fabricate 3D micro-capacitors [115] and pillared micro-reactors [116]. Electrically conductive structures have been made by 3D printing carbon nanofibres [117]. In bioengineering, applications of 3D inkjet printing range from bone and teeth printing to vascular and organ scaffold construction. Consequently, 3D inkjet printing has become an important tool for the development of biocompatible materials [118, 119] and drug delivery devices [120].

The feature of sub-micrometer accuracy and resolution makes printing technologies powerful for printing nanomaterials. The use of carbon nanotubes (CNTs) for sensors and nanoelectronics has been extensively studied. Forming Ohmic connections between CNTs and electrodes is crucial for these applications. Dockendorf *et al* [121] demonstrated the use of a fountain pen to deposit gold nanoink on top of CNTs. After solvent evaporation, the gold nanoparticles formed a conducting gold structure; thus, contacts between CNTs and electrodes were established. The fountain pen printing method was proven to produce better low contact resistances than other CNT contact formation methods. Suryavanshi and Yu [122] demonstrated a technique, which was named electrochemical fountain pen nanofabrication (ec-FPN), to deposit freestanding platinum nanowires. Similar to electro-hydrodynamic printing, ec-FPN also utilizes the meniscus shaped between an electrolyte-filled nanopipette (i.e., the fountain pen) and a substrate to dispense materials to the target substrate. In their experiment, the platinum nanowires deposited with a nanopipette, having a 100 nm aperture, had a diameter of about 150 nm and a length of 30 μm.

Printing technologies also enable the delivery of foreign materials, such as drug molecules, into cells. Using electrospinning, Zeng *et al* [123] achieved significant diameter reduction and uniformity improvement of the poly (L-lactic acid) fibre size. Inkjet printing has also been shown for biological material delivery [124]. Loh *et al* [125] reported high-resolution delivery of functionalized nanodiamonds using a nanofountain probe, via direct-write nanopatterning with a resolution of 100 nm and *in vitro* injection into cells.

Based on the advances in SAW printing, Forde *et al* [126] demonstrated the synthesis of 10 μm protein-loaded polycaprolactone polymer particles using an ultrasonic atomizer. Because the low frequencies of their method with ultrasonic atomization were insufficient to prevent protein denaturing, Mar Alvarez *et al* [54] presented high-frequency production of protein-loaded polymer microparticles and reported the effect of SAW working frequencies and polymer concentration on particle sizes.

Inkjet and e-jet printing methods have been shown to be able to print living cells. However, printing cells based on an inkjet has problems of low cell viability [127], which make it possible to repair and replace damaged/ageing tissues. Jayasinghe *et al* [128, 129] used electrospinning to form cell-bearing scaffolds that were used to directly create multi-cellular synthetic tissues and functional vessels. In addition, they have demonstrated the ability to couple gene therapy with both bio-electrosprays and cell electrospinning for the delivery of personalized therapeutics, they have also been able to engraft post-treated stem cells into mouse models [130–134].

5. Summary

In summary, a number of printing technologies are now available for generating structures at the micro and nanometer scales. Nano- and micro-scales printing techniques have found many applications in electronics, biotechnology, materials synthesis and patterning. Traditional inkjet printing has difficulties in printing sub-micrometer patterns and can cause damage to sensitive solutions. Electrohydrodynamic printing has been applied to printed electronics and biochip fabrication due to their high resolution and simple setup; however, the construction of a nozzle with high consistency and cross contamination demands further improvement. DPN and NFP have high printing resolution, high equipment requirements, and limited throughput. They have mainly been used in the precision deposition of functional materials and drug delivery. Pyroelectrodynamic printing and SAW printing are in the early phase of development. For instance, they have the advantage of simple system setup when compared to electrohydrodynamic printing, which must be equipped with physical nozzles with fine tips.

These printing technologies form a repertoire of powerful tools that enable the printing and deposition of a variety of materials, ranging from biomaterials (e.g., DNA and proteins) to nanomaterials (e.g., nanowires/tubes and nanoparticles). Printing at the micro and nanometer scales is an enabler for science and engineering research, it also has significant practical applications in manufacturing. Therefore, the next decade will witness further advances in existing printing methods and will see the emergence of new technologies.

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