Recent advances in printable carbon nanotube transistors for large-area active matrices

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ABSTRACT

Active-matrices serve as the backplane circuitry for large-area display technologies and distributed sensors. Recently, there has been significant interest in developing flexible, additively manufactured active matrices for the burgeoning flexible electronics industry. Carbon nanotubes (CNTs) are prime candidate materials for semiconducting elements of transistors due to their solution processability, carrier mobility, and mechanical flexibility. There have been many recent accomplishments in the development of CNT inks and in their deposition via printing that enable their use in thin-film transistors (TFTs), and their appropriate performance make them suitable for use in large-area active matrices. In this review, we provide an overview of the field, with a specific focus on recent advancements in CNT sorting, ink preparation, and printing techniques. We also provide a benchmarking study of printed CNT devices presented in literature after 2017. Next, we discuss printable CNT-TFTs used for active-matrix applications. Finally, we provide a concluding perspective on the outlook and challenges for printed CNT-TFT active matrices.

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1. Introduction

Active matrices allow for large-area distributed control using the minimum number of addressable connections. They are now ubiquitous in our everyday lives, enabling technologies such as displays and touch-screen sensors. In short, active matrices allow for each node to be addressed via an electronic switch. This ensures that individual nodes do not electrically affect surrounding nodes. If a system has \( m \times n \) nodes, each node can be individually addressed using only \( m + n \) electrical connections \([1]\). A passive matrix offers the same advantage in terms of connectivity, but each node is electrically coupled with the nodes on a shared row, therefore bearing significant interdependencies and cross-talk that are deleterious \([2]\). A brief schematic diagram of a common active-matrix circuit with a two-transistor, one-capacitor \((2T1C)\) architecture is shown in Figure 1A.

Active matrices were first introduced by Bernard Lechner in 1971 \([3]\) and realized in 1975 using CdSe thin-film transistors (TFTs) to address electroluminescent displays \([4]\). Since then, many other materials have been utilized for the same purpose, often with commercial success. For example, amorphous silicon can be used for low-cost applications when transistor performance is not paramount \([5]\). Conversely, Indium-Gallium-Zinc-Oxide (IGZO) provides exceptional mobility for modern high-end display technology \([6,7]\). While these two examples have matured significantly, both are rigid and rely on vacuum deposition techniques. Recently, many efforts have been made to develop both printable and flexible active-matrix circuits. A cartoon schematic of the typical device architecture, along with a direct printing method, can be seen in Figure 1B. These efforts can be broadly broken down into two categories: material advances and solution processing advances.

The primary semiconducting materials that can be utilized for flexible active electronics include metal-oxides, organic semiconductors (both conjugated polymers and small molecules), and nanomaterials. Metal-oxides often offer poor flexibility due to their ceramic nature and required high-temperature processing, but some progress has recently been made in enhancing their flexibility through advances in ultraviolet (UV) curing \([8,9]\). Conversely, organic semiconductors offer many advantages in terms of flexibility and solution processability \([10–13]\). Their primary drawback is their performance, as their...
Figure 1. Overview of the active matrix architecture and the enabling thin-film transistors (TFTs). (A) Schematic of a traditional 2T1C active matrix circuit. (B) An aerosol jet printing flexible TFT. (C) General trends in material field-effect mobility.

Mobility is drastically reduced compared to metal-oxides and nanomaterials. Nanomaterials, specifically carbon nanotubes (CNTs) but also nanomaterials comprising two-dimensional (2D) materials, offer an attractive middle ground between flexibility and performance [14,15]. For this review, we will focus on CNTs as semiconducting materials in printable active devices. A summary of the different materials, with their associated field-effect mobilities, can be found in Figure 1C.

CNTs were first discovered in 1991 [16,17], and were first introduced in transistor devices in the late 1990s [18]. They can be defined as nano-scale cylindrical carbon lattice structures that contain solely carbon atoms with sp2 bonds arranged in a hexagonal lattice. They are often described by the number of their layers—as single-walled CNTs (SWNTs), few-walled CNTs, and multi-walled CNTs (MWNTs)—and their electronic properties are determined by the chirality of their tube [19]. The band structure of a CNT can be determined by analyzing the quantization of the graphene band structure due to the confinement that occurs when the lattice exists in the nanoscale cylindrical shape [20].

For printable devices extending from CNTs, the semiconducting thin-film occurs as a network of many individual CNTs [21]. This network is typically randomized, as seen in Figure 2A. The randomness of the interconnections averages out the material property, and the resulting electrical properties can be determined by the individual electronic properties of the CNTs, the junction electronic properties, and the overall density of the CNTs. In 2001, McEuen et al. [24] were able to individually probe CNT junctions, and they experimentally demonstrated

Figure 2. Electrical properties of randomized CNT thin-film networks. (A) Scanning electron microscopy of a printed randomized network. Adapted with permission from [23], copyright 2016, American Chemical Society. (B) Plot of the field-effect mobility with respect to the networked tube density. (C) Plot of the field-effect mobility with respect to the on/off ratio of the transistor. (B) and (C) are adapted with permission from [22], copyright 2010, John Wiley and Sons.
that the junction between two semiconducting CNTs exhibits ohmic behavior, with a nominal resistance value of approximately. To decrease the sheet resistance of networked CNT films within the on-state, it is important to have many of these high-resistance junctions parallel to one another. Therefore, there is a direct relationship between the CNT density and the device mobility. The higher the density of the tubes is, however, the higher the overall off-current of the device becomes, particularly if the purity of the nanotubes used is not ideal. The relationship between the density and the mobility, as well as the trade-off between the density and the on/off ratio, was demonstrated by Burke et al. [22] and is shown in Figure 2B and C. Overall, networked CNT devices exhibit a semiconducting behavior with effective mobility values that range from 1 to 80 cm²/Vs. This, coupled with their printability, make them an attractive alternative to organic semiconductors for developing printable TFTs used for active matrices.

In this review, we will focus on three areas of interest for CNT-TFT active matrices – CNT processing and deposition, CNT-TFT performance, and CNT-TFT integration into active matrices for various applications. For processing, we will briefly cover methods of synthesizing CNTs, followed by in-depth descriptions of sorting techniques for generating CNT inks. This will be followed by a review of recent advances in CNT deposition, namely through direct-write printing approaches such as aerosol jet printing and inkjet printing. Next, we will benchmark the performance of various CNT-TFTs presented in literature. Finally, we will discuss the latest demonstrations of the use of CNT-TFTs for active-matrix applications, specifically for displays and AM-enabled distributed sensors.

2. CNT processing

2.1. Synthesis of CNTs

CNTs were first synthesized in 1991 when Iijima et al. [16] reported the formation of tubular needles through the arc-discharge evaporation method. Very simply, the arc-discharge process brings together two electrodes, an anode and a cathode, with a power supply to generate an arc [25]. The electrodes are typically composed of pure graphite, and the anode can be optionally loaded with a carbon precursor and catalyst. The arc that occurs between the electrodes forms a plasma, which promotes sublimation of the anode; and subsequently, the carbon vapor is deposited on the cathode in the form of a mixture of nanotubes and amorphous carbon [26]. The current, catalyst, temperature, and inert gas used in the reaction chamber will determine the type of CNT: either MWNT or SWNT [27]. SWNTs are a graphene sheet rolled to form a tubular structure, and MWNTs are a series of rolled concentric graphene sheets [28]. The tubular needles that Iijima et al. [16] synthesized were MWNTs made from a pure graphite anode and cathode in a helium environment.

In the mid-1990s, the arc-discharge method had challenges in synthesizing high-quality and uniform CNTs, specifically SWNTs [27]. This led to the development of two other main synthesis strategies: laser ablation [29] and chemical vapor deposition (CVD) [30]. The laser ablation technique involves the vaporization of graphite rods (with or without a catalyst) with a continuous or pulse laser source [27,29]. The CVD process generally involves catalyst-assisted decomposition of hydrocarbons that, after being cooled in a tube reactor, form CNTs [31]. Unlike the arc discharge and laser ablation processes, CVD allows for greater control of the chirality and diameter of the synthesized CNTs without additional purification [32]. This has made CVD techniques, such as those that use high-pressure carbon monoxide (HiPCO) and a cobalt molybdenum catalyst (CoMoCAT), the industry standards for SWNT synthesis.

2.2. Sorting of CNTs

The critical drawbacks of the aforementioned synthesis techniques are the overall lack of quality, yield, and uniformity of the CNTs. Specific synthesis techniques can produce CNTs with impurities, amorphous carbon, and a significant defect density. Moreover, the produced CNTs are typically composed of nanotubes with different diameters and electronic types (metallic or semiconducting) [33]. In CNT-TFTs, the current on/off ratio (I_on/off) is crucial in the performance of the transistor device. In the presence of metallic conductive tubes, the I_on/off of the device will be minimal [34]. Notably, in metallic and semiconducting CNTs, the dominant optical transitions vary with the chiral vector and diameter of the nanotube [35]. In addition, the bandgap of semiconducting CNTs varies inversely with its diameter [35]. Thus, sorting strategies for semiconducting CNTs are primarily driven by the separation of the tubes based on their chirality to achieve a monodispersed single chirality population. The four primary methods used are density gradient ultracentrifugation (DGU) [36], gel chromatography [37], the aqueous two-phase system (ATPS) [38], and conjugated polymer wrapping [39].

In 2005, Arnold et al. [36] discovered that the DGU separation technique could be used to sort CNTs. DGU combines specific surfactants into the CNT solution; and when a high centripetal force is administered in a density medium, the solution separates into discrete
bands based on their buoyant density [36,40,41]. The resulting supernatant allows for selective separation of CNT species based on their chirality. The process has been shown to significantly refine the diameter within arc-discharge CNTs [42]. These early results were auspicious, and the discovery that surfactant coatings are correlated with the electrical properties of CNTs led to the maturation of the gel chromatography and ATPS techniques [43].

In 2009, Tanaka et al. [37] reported a simple procedure for achieving a 95%-containing semiconducting SWNT gel resulting from gel squeezing. The agarose-CNT gel was frozen, thawed, and squeezed to remove metallic CNTs from the gel. Shortly after, Moshammer et al. [44] conducted a chromatography separation by pouring the CNT solution through a polysaccharide gel column. The technique has since adopted the single-surfactant gel multicolumn method, which involves several gel columns connected in series [45,46]. This method allows for large-scale chirality sorting with the use of a single surfactant. Recently, due to the low flow rate of CNT solutions through gel columns, Khamidullin et al. [47] presented the use of chemically modified cotton wool as a cost-effective replacement to polysaccharide gels.

ATPS was first utilized by Khripin et al. [38] in 2013 to sort CNTs. They observed the spontaneous CNT partition when ATPS was added to two immiscible polymers. The authors found that the metallic CNTs separated into the more hydrophilic phase, and the semiconducting CNTs moved into the hydrophobic phase [38]. An issue with CNT sorting via ATPS is the time-consuming multistep process of sorting particular CNT species one by one [48]. Podlesny et al. [48] used basic salts to modulate the CNT partitioning in order to reduce the number of steps. Karandish et al. [49] went further by demonstrating that one of the polymer phases of the ATPS could be substituted with a salt, such as sodium citrate, to lower the cost, and the ATPS would still be capable of portioning SWNTs.

The use of a conjugated polymer in CNT wrapping was first reported in 1998 by Curran et al. [50]. They reported that poly(p-phenylene vinylene) helped disperse MWNTs in the polymer solution. The mechanism behind this phenomenon is well understood as the noncovalent functionalization of CNTs, which involves the wrapping of the polymer chain around the external nanotube sidewall through Vander Waals interactions [51,52]. The side chains of the wrapped polymer allow for dispersion of the SWNTs in a solvent. It was not until 2007 when Nish et al. [39] reported that polyfluorene-based copolymers were capable of purifying SWNTs to a single species and to the desired chirality. Since then, more conjugated polymers have been realized in the sorting of semiconducting SWNTs, such as polycarbazoles, polythiophenes, and donor-acceptor polymers [53]. Compared to other sorting techniques mentioned here, conjugated polymer wrapping has received considerable attention due to the relative simplicity of its process and its capability to produce high-purity CNTs. The one-pot separation technique discussed by Ozawa et al. [54] highlights the simplicity of the process where a SWNT-conjugated polymer solution is sonicated, then centrifuged. Figure 3 illustrates the typical semiconducting CNT sorting procedure in polymer wrapping. After centrifugation, the supernatant, which is composed of the separated semiconducting CNTs, is extracted. An unfortunate drawback to polymer wrapping is the presence of excess polymer that is not bound to the CNTs and is then deposited alongside the CNTs. In CNT-TFTs, the inclusion of polymer in the SWNT active channel can hinder the electrical transport of the device [51]. Typically, the excess polymer must be repeatedly filtered and rinsed to maximize the device performance.

Regardless of the sorting strategy implemented, once the desired purity is reached, the next phase is to determine the method of CNT deposition. An evolving alternative to industry-standard photolithography CNT
deposition techniques is printing technologies. The following section describes various printing techniques that are used for CNT-TFT devices.

After the CNTs are sorted, inks are formulated in different ways. The two main considerations for the selection of the ink formulation method are the CNT dispersion stability and compatibility with the subsequent printing process. For many applications, the sorting polymer can also be used as the dispersion agent. For example, CNTs that are sorted using polyfluorene-based co-polymers can be dispersed in aromatic solvents to provide shelf-stable dispersions for up to six months [55]. Another method of CNT dispersion is to utilize a small molecule surfactant, such as sodium dodecylsulfate, in aqueous solutions [56].

Once the dispersion method is identified, the solutions can be further modified to make them compatible with the desired printing process. For inkjet printing, where viscosities close to 10 cP are desired, various co-solvents, such as N-Cyclohexyl-2-pyrrolidone (CHP), can be utilized in conjunction with aqueous or aromatic solutions to increase the viscosity [57]. For aerosol jet printing, there have been demonstrations of direct printing of a diluted version of the dispersed ink [58], as well as demonstrations of increased printing stability after the addition of a viscous co-solvent, terpinol [59].

3. Printing techniques

Printed electronics is a rapidly expanding field, with numerous techniques such as aerosol jet printing and inkjet printing being investigated for novel applications. One of these applications is the printing of active devices, such as CNT-TFTs [59,60]. Printing, or the controlled additive deposition of electronic inks, offers many cited advantages such as cost [61], sustainability [62], and compatibility with flexible/stretchable substrates [63]. In this paper, we discuss recent advances and challenges in printing techniques for CNT-TFTs.

3.1. Aerosol jet printing

Aerosol jet printing has been widely used to print CNTs due to its compatibility with different nanomaterial solutions of varying viscosity and its capability to pattern fine circuit structures with line widths as small as 10 microns [64]. In an aerosol jet printing system, CNTs dispersed in solutions are atomized via ultrasonication or pneumatically. The atomized aerosol droplets are then carried by N$_2$ gas flow to the nozzle, where they are focused by the sheath gas flow and deposited onto the substrate. The sheath gas flow serves two purposes: to aerodynamically focus the ink and prevent nozzle clogging by ensuring that the ink will not come into contact with the nozzle sidewall. The key process parameters for aerosol jet printing are the atomization parameters (the ultrasonicator current or the pneumatic flow rate), the carrier gas flow rate, the sheath gas flow rate, the nozzle size, and the platen temperature. These parameters need to be optimized for specific materials [65]. A schematic overview of the ultrasonic atomization to deposition process flow for aerosol jet printing is shown in Figure 4A.

In the early stage of its development, aerosol jet printing was used to deposit only CNTs as active channel materials. Source, drain, and gate electrodes were deposited using photolithography, electron-beam lithography (EBL), and thermal evaporation [14,59,67,68]. Although this strategy is good for achieving a uniform device geometry and reducing the number of variables to characterize the electrical transport properties of CNT-TFTs, a fully printable device must be realized to obtain the many cited advantages of printed electronics.

Another important process in the fabrication of CNT-TFTs is the deposition of their dielectric layer. Atomic layer deposition (ALD) and thermal evaporation have been used to deposit materials with a high dielectric constant such as HfO$_x$ [59] and AlO$_x$ [67]. Although these techniques are effective in producing ultrathin and pinhole-free dielectric layers, they are not compatible with low-cost printing techniques. To overcome this challenge, printable dielectrics have been the focus of research. Printable ion-gel dielectrics [69] and polyfluorinated electrolytes [70] have shown great potential for complete device integration.

Recently, fully aerosol jet-printed CNT devices have been demonstrated, and many advancements have been made [58,71,72]. For example, Lu et al. [73] improved the uniformity and stability of aerosol jet-printed CNT-TFTs by controlling the ink temperature within the sonication bath. The study presented a significantly reduced standard deviation in terms of an effective mobility of only 4%. Other studies looked specifically at the contact materials utilized in printable CNT-TFTs. Cardenas et al. [74] studied the impact of the morphology of the printed contacts on the performance of aerosol jet-printed CNT-TFTs. They showed that electrodes printed with silver nanoflakes provide the best electrical interfaces. Andrews et al. [75] explored the possibility of using printed liquid metals as contacts for CNT-TFTs. They showed that although liquid metal contacts produced with direct writing techniques have higher contact resistance than liquid metal-filled PDMS microchannel contacts, liquid metal contacts could open a path for fully stretchable CNT-TFT devices.

Although aerosol jet printing has shown great promise for fabricating CNT-TFT devices, it still faces challenges.
An intrinsic drawback of aerosol jet printing is the overspray, which results in poorly defined patterns [76]. The gradual change of the CNT ink density during the extended printing period and the long-term on-shelf stability of CNT inks can be present challenges in controlling the device-to-device uniformity [59].

### 3.2. Inkjet printing

Inkjet printing is a drop-on-demand technique, where material inks are deposited onto substrates with the help of the piezoelectric response of nozzle heads. Inkjet printing systems are highly sensitive to the viscosity of the material inks due to the capillary mechanism of deposition. However, the technique is efficient in printing large-area electronics with decent patterning accuracy. The key printing parameters for inkjet printing are the ink viscosity and the surface tension, drop spacing, jetting voltage, drop speed, and platen temperature. Matching the ink fluid properties with the surface properties of the substrate is critical for successful printing [60]. A schematic of inkjet printing CNT-TFTs is shown in Figure 4B.

Similar to aerosol jet printing, inkjet printing has been used to deposit CNTs to the channel region of the device. Historically, inkjet-printed CNT-TFTs have performed poorly because of the high metallic CNT content of the ink solution. With the development of new sorting techniques and commercially available high-purity semiconducting CNT inks (as discussed in section 2 of this review), the performance of inkjet-printed CNT-TFTs has improved significantly. Bucella et al. [77] demonstrated high-performance complementary logic circuits fabricated with inkjet printing CNTs. They also showed that CNT inks with densities higher than 0.2 mg/L clog nozzles, so an upper density limit for printable inks is set. Remarkably, a single drop of inkjet-deposited CNT ink has been used as an active channel material in multiple studies [78,79], which demonstrates the material cost-effectiveness of the inkjet printing method.

Yoo et al. [80] presented a fully printed double-gated CNT-TFT. By allowing a dual-gate structure, they were able to minimize the threshold voltage shift in fully printed CNT-TFTs over time. Lee et al. [81] demonstrated fully inkjet-printed CNT-TFTs with improved contact interfaces. They reduced the contact resistance by increasing the CNT network densities under the source and drain electrodes. Since in inkjet printing, the ink comes in direct contact with the sidewalls of the cartridge, nozzle clogging is a serious issue, unlike in aerosol jet printing. Sufficient dispersion of the CNTs and filtering out of large, agglomerated particles is essential for successful inkjet printing [82].

Both aerosol jet printing and inkjet printing are known to suffer from coffee ring effects, which are due to the replenishment of the ink droplet solvent that evaporated from the edge with the solvent from the center of the droplet. However, researchers have used the coffee ring effect to their benefit and explored the possibility of printing self-aligned CNT twin lines [83], which could be exploited to improve the transport performance of CNT network transistors. Cao et al. [84] demonstrated a printed submicron channel-length CNT transistor by taking advantage of the hydrophobic repulsion between the SAM layer and the gold ink. Song et al. [85] developed a process called SCALE (self-aligned capillarity-assisted lithography for electronics) to take advantage of the capillary force to self-align the different inkjet-printed components of CNT-TFTs.

![Figure 4](image_url)  
*Figure 4. Schematics of different printing techniques: (a) aerosol jet printing, (b) inkjet printing, and (c) roll-to-roll (R2R) printing. Figure (c) is adapted from [66], copyright 2012, Elsevier.*
3.3. Other printing techniques

Direct-write printing techniques, such as the previously described aerosol jet printing and inkjet printing techniques, are good for early-stage research and development of CNT-TFT-based devices. However, for large-area, mass-scale production, roll-to-roll (R2R) printing techniques, such as gravure printing [86], roll-to-plate printing [87], screen printing [88], spray printing [89], or a combination of these techniques, will be required. Figure 4C illustrates different R2R printing techniques. Although these techniques require different templates for different circuit patterns, the cost of the templates could be significantly mitigated through mass-scale production. To date, printing CNT thin-films using R2R techniques is incredibly challenging due to the viscosity requirements and the lack of stable CNT solutions. However, a recent study used the novel ligand exchange technique to enable R2R printing of CNT-TFTs [90]. The study is discussed further in the Application section, as it uses the technique specifically for active-matrix applications.

4. CNT-TFT performance

Solution-processed TFTs are transistors composed of a semiconducting layer, an insulator/dielectric layer, and three electrodes (the source, drain, and gate electrodes). A primary difference between these thin-film devices and the traditional metal–oxide-semiconductor field-effect transistors (MOSFETs) is the underlying bulk Si body effect [91]. While all layers are crucial, the specific arrangement of layers and electrodes can be modified and defines the architecture of the TFT. The four classical configurations of solution-processed TFTs (shown in Figure 5) are bottom gate/bottom contact, bottom gate/top contact, top gate/bottom contact, and top gate/top contact. Bottom-gate CNT-TFTs are semiconducting SWNTs deposited on the gate/dielectric and connected to the metallic source and drain contacts. The bottom/top contact nomenclature signifies the order in which the SWNTs are deposited: before or after the source and drain electrodes.

When a voltage bias is applied between the source and drain electrodes, the resulting current that flows through the semiconducting layer depends heavily on the bias voltage applied to the gate electrode. The mechanism behind this coupling is the lowering of the Schottky energy barrier by the gate field to increase the probability that charge carriers will tunnel through the semiconducting CNT network [34]. This implies that when no gate voltage is applied to the CNT-TFT, the mobility of the charge carriers is limited and very low [92]. Thus, essential metrics, such as $I_{on}$ and $I_{off}$, can be derived to represent the on-state current and the off-state current. The ratio of the on-state current to the off-state ($I_{on}/I_{off}$) current is used to evaluate and benchmark the performance of CNT-TFT devices. For large-area active matrices, a low

![Figure 5. Illustration of the four common TFT device architectures.](image-url)
$I_{\text{off}}$ and a large $I_{\text{on}}/I_{\text{off}}$ are sought after to promote low power consumption and display quality [93]. Another important parameter for the characterization of CNT-TFTs is the charge carrier mobility—the average charge carrier drift velocity under an applied electric field. Simply, this measures how efficiently the charge carriers can flow within the CNT-TFT. The mobility can be determined from the transfer characteristics in the linear or saturation regions [94]. The mobility determined in the linear region is referred to as the effective mobility ($\mu$), which is calculated as follows [34]:

$$\mu = \frac{L_{\text{CH}}g_m}{W_{\text{CH}}C_{\text{OX}}V_{DS}}$$

where $L_{\text{CH}}$ is the channel length, $W_{\text{CH}}$ is the channel width, $g_m$ is the peak transconductance, $C_{\text{OX}}$ is the gate capacitance per unit area, and $V_{DS}$ is the applied drain–source voltage. As mentioned, the CNT density and the defect density play a critical role in achieving higher mobilities but often at the expense of a larger $I_{\text{off}}$. CNT-TFTs can exhibit an $I_{\text{on}}/I_{\text{off}}$ ratio of up to $10^7$ and a mobility of up to 90 cm$^2$/Vs [95,96].

Typically for printed semiconducting SWNT layers, the CNT networks are classified as random networks. CNT-TFTs with random networks normally have mobilities that are much smaller than those of comparable CNT-TFTs with aligned networks [97]. As mentioned, researchers have manipulated the deposition and evaporation rates of CNT inks during printing to achieve CNT alignment. Combining aligned CNTs with a low-cost and flexible substrate deposition process would improve the quality of large-area active matrices.

Recent studies (2017–2021) were compiled to benchmark the performance of printed CNT-TFTs. To be included in this analysis, the semiconducting CNT layer had to be deposited via a printing technique (e.g. aerosol or inkjet). Figure 6 describes the mobility versus $I_{\text{on}}/I_{\text{off}}$ ratios of the fabricated CNT-TFT devices in each selected research article. The corresponding table, Table 1, provides further details for each device. Interestingly, it shows a shift from inkjet printing of semiconducting inks to aerosol jet printing over time. This shift can be attributed to the increased flexibility in ink preparation with aerosol jet printing. As seen in Figure 6, the CNT-TFT reported in article [96] exhibited significant mobility, 90 cm$^2$/Vs, nearly double the mobility of any reported printed CNT-TFT device. This increased mobility resulted in part from the SWNT alignment in the active channel after inkjet printing. The mechanism of the alignment was attributed to the SWNT interaction with an octadecyltrichlorosilane-modified SiO$_2$ dielectric layer [96]. This research highlights the potential of aligned CNT printing to develop high-performance CNT-TFTs.

### 5. Applications

The primary applications explored using printable CNT-TFTs in recent literature include circuitry [112–115], sensors [72,116], and active matrices [90,117–120]. The efforts at achieving appropriate properties for facilitating

![Figure 6. Mobility versus the $I_{\text{on}}/I_{\text{off}}$ ratio of CNT-TFT devices with printed s-SWNT layers.](image-url)
digital logic or other rudimentary circuit applications are not promising, as the mobility is low and the effective area for printable CNT-TFTs is large. However, for generating supporting elements within an active-RFID technology, the results can be quite promising. Jung et al. [121] used p-type CNT-TFTs to generate a cyclic waveform from a DC voltage. This allowed them to integrate the circuitry into a fully functional cyclic voltammetry sensing device in a few printing steps. Ultimately, their study demonstrated that rudimentary TFTs can be printed to enable a novel device while increasing its manufacturability.

Even more recently, innovations have been made to allow for n-type performance of printed CNTs [105]. These have led to the introduction of circuitry compatible with the complementary metal–oxide semiconductor (CMOS). Gao et al. [111] developed an example of a printable CMOS-compatible CNT inverter that uses a composite polymer solid-state electrolyte (CPSE) dielectric material to enable printable CNT-TFTs with a low operating voltage. Specifically, they used a crosslinked poly(4-vinylphenol) doped with an ionic liquid to form the CPSE. They were able to achieve both the n- and the p-type, with the n-type requiring an additional epoxy amine ink layer. Using the complementary transistors, they fabricated a two-transistor inverter. The inverter was capable of full rail-to-rail voltage operations at \( V_{DD} \) values down to 0.5 V. This study represents a promising step towards the development of fully printable complementary gates, which could potentially enable on-board CMOS logic.

Along with circuitry, electronic transduction elements from printable CNT-TFTs for sensing applications have also been developed, with significant advantages noted. These include physical transducers for sensing pressure, strain, and temperature. A fully printable CNT-TFT was shown to have a transconductance that was linearly related to environmental pressure over a wide sensing range [72]. The schematic can be seen in Figure 7. The linear relationship to environmental pressure could stem from three main mechanisms proposed by the authors: (1) the pressure reducing the sheet resistance of the nanotube thin-film, (2) the pressure-reduced thin contact resistance of the film, or (3) the dielectric film deformation leading to an increased gate capacitance. Additionally, CNT-TFTs are extensively discussed in literature for demonstrations of both chemical and biological sensing. Some recent efforts include utilizing CNT-TFTs for sensing nitrogen-based gases such as ammonia and nitrite, and incorporating CNT-TFT transducers into traditional sandwich-based immunoassays for electrical detection of biomarkers. A more extensive review of advances in this area can be found in Schroeder et al. [122].

However, the focus of this review is CNT-TFTs within active-matrix applications. The two main applications of these active matrices, which have been discussed in previous literature, are applications based in displays and those for handling distributed sensors. Utilizing printable CNT-TFTs for sensing applications provides a communication structure capable of handling large amounts of distributed sensors over large areas in a flexible form factor.

Active matrix-enabled distributed sensing platforms have been demonstrated in conjunction with pressure sensors, which could have had an impact in the soft

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**Table 1. Benchmark comparison of printed s-SWNT layers in CNT-TFTs. PW refers to ‘polymer wrapping.’**

| Year          | Process technique | SWNT type            | Sorting      | Application          | \( \text{log}(I_p/I_{th}) \) | Mobility (\text{cm}^2/\text{V}s) | Citation |
|---------------|-------------------|-----------------------|--------------|----------------------|-----------------------------|---------------------------------|----------|
| 2017          | Inkjet Printing   | Proprietary           | PW: Proprietary | Flexible TFT        | 4.00                        | 75.0                            | [85]     |
| 2017          | Inkjet Printing   | Arc Discharge         | PW: Polyfluorene-based | Non-specific TFT   | 7.48                        | 30.0                            | [95]     |
| 2017          | Inkjet Printing   | HiPco                 | Gel Chromatography | Non-specific TFT   | 6.00                        | 90.0                            | [96]     |
| 2017          | Inkjet Printing   | Arc Discharge         | PW: Oligothiophene-based | Non-specific TFT   | 6.00                        | 57.0                            | [98]     |
| 2017          | Inkjet Printing   | Proprietary           | PW: Proprietary | Non-specific TFT    | 5.00                        | 15.03                           | [84]     |
| 2017          | Inkjet Printing   | Proprietary           | PW: Proprietary | Flexible TFT        | 6.00                        | 6.00                            | [57]     |
| 2018          | Aerosol Jet Printing | Proprietary          | PW: Proprietary | Non-specific TFT    | 5.00                        | 12.25                           | [99]     |
| 2018          | Inkjet Printing   | Proprietary           | PW: Proprietary | CMOS inverter       | 2.43                        | 0.24                            | [100]    |
| 2019          | Aerosol Jet Printing | Proprietary          | PW: Proprietary | Flexible TFT        | 5.54                        | 10.20                           | [71]     |
| 2019          | Inkjet Printing   | Plasma                | PW: Polyfluorene-based | Flexible TFT       | 4.00                        | 27.00                           | [101]    |
| 2019          | Aerosol Jet Printing | Arc Discharge        | PW: Polyfluorene-based | Non-specific TFT   | 5.00                        | 11.00                           | [102]    |
| 2020          | Aerosol Jet Printing | Proprietary          | PW: Proprietary | Flexible TFT        | 6.00                        | 8.00                            | [103]    |
| 2020          | Inkjet Printing   | Proprietary           | PW: Proprietary | Non-specific TFT    | -                           | 12.03                           | [80]     |
| 2020          | Aerosol Jet Printing | Arc Discharge        | PW: Polycarbazole | Gas sensor          | 5.00                        | 9.90                            | [104]    |
| 2020          | Aerosol Jet Printing | Arc Discharge        | PW: Polyfluorene-based | CMOS inverter     | 6.00                        | 8.90                            | [105]    |
| 2020          | Roll to Roll Printing | Plasma              | PW: Polyfluorene-based | Active matrix     | 4.10                        | 0.23                            | [90]     |
| 2020          | Aerosol Jet Printing | Arc Discharge        | PW: Polyfluorene-based | CMOS inverter     | –                           | 8.00                            | [67]     |
| 2021          | Aerosol Jet Printing | Proprietary          | PW: Proprietary | Flexible TFT        | 3.00                        | 10.00                           | [106]    |
| 2021          | Aerosol Jet Printing | Arc Discharge        | None          | Flexible TFT        | 6.48                        | 2.50                            | [107]    |
| 2021          | Aerosol Jet Printing | Arc Discharge        | PW: Polyfluorene-based | Gas sensor        | 6.00                        | 9.80                            | [108]    |
| 2021          | Aerosol Jet Printing | Arc Discharge        | PW: Polyfluorene-based | Non-specific TFT  | 5.10                        | 10.80                           | [109]    |
| 2021          | Aerosol Jet Printing | Arc Discharge        | PW: Polyfluorene-based | CMOS inverter     | 5.30                        | 8.20                            | [110]    |
| 2021          | Aerosol Jet Printing | Arc Discharge        | PW: Polyfluorene-based | CMOS inverter     | 6.00                        | 6.60                            | [111]    |
Figure 7. Fully printable CNT-TFT used as an electronic environmental pressure transducer. (A) The device being flexed. (B) Schematic of the backgated, top contact-printed CNT-TFT. (C) Transconductance with respect to environmental pressure inside a pressurized chamber. The figure is adapted from [72], copyright 2018, Institute of Electrical and Electronics Engineers.

robotics space. In 2016, Yeom et al. [117] demonstrated a 45 cm² backplane fabricated using a combination of R2R printing and other solution processing techniques. They used CNT-TFTs to interrogate an array of pressure-sensitive rubber (PSR) nodes, and were able to demonstrate individual pixel addressability through the active-matrix architecture. A significant issue that they were able to overcome in their study is device-to-device variability. They were able to show a 97% yield from 400 individual TFTs with limited variability in their on-current (4 ± 2 μA/mm) and on/off ratios (4 ± 0.4 decades). They also demonstrated that their CNT-TFTs showed little degradation due to strain, which opened up the possibility for flexible or stretchable applications.

In other studies, Nela et al. [118] demonstrated a non-printed yet still solution-processed CNT-TFT array connected to (PSR) nodes. Their device consisted of 16 × 16...
Figure 9. A fully R2R-printed CNT-TFT active matrix to enable an electrophoretic display. (A) The R2R process with the circuit schematic and layer diagram of the device. (B) Plot of the performance of 400 devices with the inset elaborating the yield. (C) The CNT-TFT active matrix-controlled electrophoretic display operating while flexed. The figure is adapted with permission from [90], copyright 2020, John Wiley and Sons.

pixels that spanned a 4-inch area and was able to operate on a small voltage range of 3 V. Additionally, the active matrix design allowed for a high sensor response rate of < 30 ms. A picture of the device, along with an optical micrograph and the results of the spatial sensing experiment, can be seen in Figure 8.

In addition to sensing applications, printable CNT-TFTs have recently been demonstrated for the enabling of active-matrix displays. A notable example is a CNT-TFT active matrix used to control a polymer-dispersed liquid crystal display [120]. The demonstration took advantage of the high on/off ratio of the CNT-TFT and of the transparency of CNT thin films. Through the use of the materials, fully flexible and transparent display backplanes were demonstrated.

The final recent notable application is the fully printed CNT-TFT active matrix demonstrated by Sun et al. [90] in 2020. In their study, they formulated a novel hydrophilic gravure-compatible ink from high-purity semiconducting CNTs using the ligand exchange method. This step allowed a full system to be fabricated in which each layer was deposited via R2R printing. An active-matrix backplane was achieved where CNT-TFT transistors controlled electrophoretic pixels at a density of 10–40 PPI. The device also showed a low threshold voltage variation of only ±13% — a key metric that allowed for uniform addressability of pixels. An overview of the R2R process and the resulting circuitry can be seen in Figure 9A, along with a summation of the device performance (Figure 9B) and an image of the electrophoretic display being bent during operation (Figure 9C).

6. Conclusion and future outlook

In only a few decades, CNTs have emerged as viable candidate materials for enabling next-generation printable and flexible active matrices. In particular, the ease of
solution processability, enabled by solution-phase sorting techniques, coupled with the emergence of nanomaterial-based printing methods, have allowed recent exploration of viable devices with impressive performance. In this review, we outlined the complete process from raw materials to devices, including CNT synthesis procedures, electronic purity sorting methods, ink development techniques, printing approaches, and the resulting device performance benchmarks.

While the strides are evident, many challenges remain that must be overcome to allow for these printable active matrices to transition out of the research laboratory. One specific challenge that stands out is the device-to-device variability exhibited by printable CNT-TFTs with respect to both the channel sheet resistance and the threshold voltage. The percolation-based carrier transport is highly dependent on the density of the CNT network; therefore, printing methods need to be controlled to maintain low variations in the CNT density over long printing periods [59]. While passive methods of controlling the CNT uniformity have been identified [73], a potentially transformative technique is closed-loop monitoring. It has been recently demonstrated for aerosol jet-printed graphene thin films [123], but has yet to be validated with semiconducting CNTs. Additionally, for active matrices to be effective, uniform threshold voltage between devices is required. CNT-TFT threshold voltages can vary strongly due to environmental conditions and substrate interactions. Therefore, care will have to be taken in developing appropriate substrate functionalization methods in conjunction with passivation schemes for the devices to transition out of the laboratory.

Another related and notable missing investigation from recent literature involves environmental resiliency testing. Many demonstrations are completed in laboratory environments with controlled humidity and temperature. Better understanding is needed of how CNT-TFTs, specifically active-matrix circuitry that contain CNT-TFTs, perform over extended periods in environmental test chambers. Such investigation could shed light on the applicability of CNT-TFT active matrices in harsh environment scenarios.

Active matrices enabled by printed CNT-TFT technology present an interesting intersection between a promising technology and significant unaddressed challenges. This strongly drives continued research on the subject while being mindful that a more focused approach directed towards the outstanding challenges is required. Once uniformity issues are addressed and environmental resiliency questions are answered, printable active matrices enabled through printable CNT-TFTs can impact many fields, including flexible display technologies and sensing backplanes.

Disclosure statement
No potential conflict of interest was reported by the authors.

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