Liquid Crystal Displays with Printed Carbon-based Recyclable Transistor Backplanes

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***Abstract*—We report the first demonstration of displays driven by embedded transistors that were additively manufactured entirely by aerosol jet printing. The backplanes of the liquid crystal displays (LCDs) consist of transistors printed from graphene, carbon nanotubes, and crystalline nanocellulose onto a glass substrate with prepatterned indium tin oxide electrodes. We addressed challenges of integrating fully printed devices into both the crossbar array structure and layered vertical structure required for an LCD, showing successful pixel switching at up to 60 Hz. As these thin-film transistors are printed exclusively from carbon-based recyclable materials, without high temperatures or vacuum processing, they offer a promising means for reducing waste in future display technologies.**

***Index Terms*—Aerosol jet printing, carbon nanotubes, crystalline nanocellulose, graphene, liquid crystal display, recyclable electronics, thin-film transistor.**

I. Introduction

Electronic displays, such as liquid crystal displays (LCDs) and organic light-emitting diode (OLED) displays are ubiquitous. Inside each display is a grid of interconnected thin-film transistors (TFTs), with one or more TFT located at each pixel to regulate its brightness. In modern displays this control circuitry, known collectively as the display backplane, is most commonly composed of TFTs using low-temperature polycrystalline silicon (LTPS) or metal oxide (e.g., indium gallium zinc oxide, IGZO) as the semiconducting channel [1]. However, TFTs from these materials have some disadvantages, including significant environmental impact in their manufacture [2]-[7]. Two of the foremost challenges contributing to the large environmental footprint of incumbent TFT technologies are: 1) the use of vacuum-based processing with hazardous gases leading to the emission of fluorinated greenhouse gases (F-GHGs), and 2) energy usage of the manufacturing equipment, which has increased by orders of magnitude over the past few decades [8]. These issues have led to extensive research into alternative materials and manufacturing approaches for TFTs used in displays [9]-[14].

One such alternative is based on a thin film of semiconducting carbon nanotubes (CNTs) [15]-[17], from which we recently demonstrated fully printed and recyclable TFTs using crystalline nanocellulose (CNC) dielectrics [18]. There are a few other reports of recyclable devices, including Park *et al.* using an organic semiconductor to create devices for wearables [19], [20]. Applying one of these recyclable device technologies to display backplanes using additive manufacturing techniques would be transformative, but the significant obstacles to integrating a fully printed TFT with an active display have precluded advancement in this direction.

Still, foundational studies on discrete TFT circuits driving single pixels [9]-[14] have led to impressive demonstrations of electronic displays driven by alternative semiconductor materials, including pentacene [21], indium oxide nanowires [22], [23], and CNTs [24]-[28]. Several works even leveraged emerging materials printing methods such as inkjet [29]-[31] and screen printing [32]-[34]. However, in most cases, finishing these TFT arrays relied on at least one traditional vacuum-based fabrication technique (vapor deposition, plasma etching, etc.), highlighting how difficult it can be to replace the traditional manufacturing of incumbent technologies with additive manufacturing techniques.

In this work, LCDs controlled by an active matrix of all-carbon printed TFTs are demonstrated. A device encapsulation scheme was developed for protecting the fully printed TFTs from resolubilizing while integrated into the layered-vertical and crossbar-array structures of LCDs. This advance enabled, for the first time, the fabrication of TFTs integrated into a display using only additive manufacturing and recyclable carbon-based electronic materials.

II. Device Fabrication

Fabrication began on polished soda lime glass substrates (Lumtec, LT-G001, 50 mm × 50 mm, 0.7 mm thick). These substrates came coated with 120-160 nm of indium tin oxide (ITO) on one side (~15 Ω/sq and >84% optical transparency). The ITO electrode geometry was defined with photolithography (Fig. 1a) and soaking in ITO etchant (Transene, TE-100) for 15 min at 60 °C. The substrates were rinsed in deionized water, acetone, and isopropyl alcohol to

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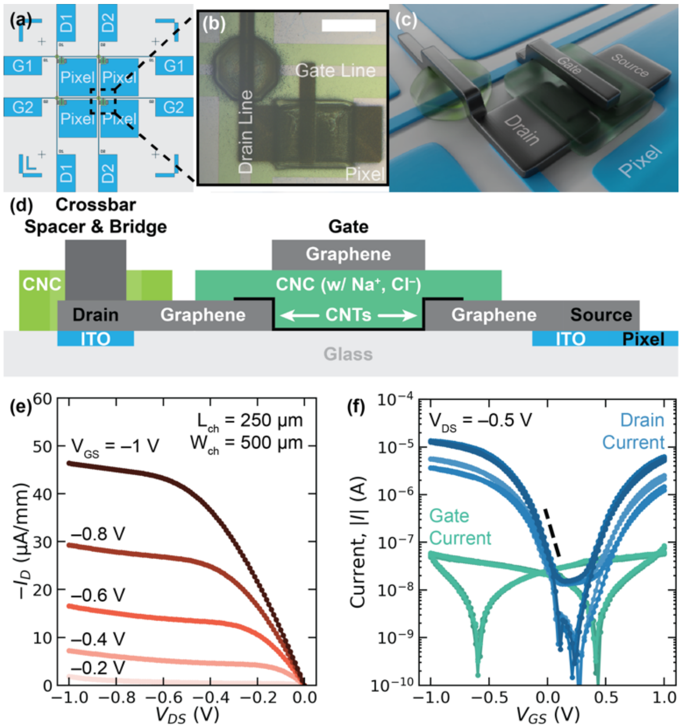

Fig. 1. Integration of all-carbon recyclable TFTs as backplanes for LCDs, including structure and TFT characterization. (**a**) Top-down schematic of 2×2 pixel array, with large ITO features labeled. (**b**) Optical image of TFT fully printed from all carbon-based materials. Scale bar is 1 mm. (**c**) Three-dimensional rendering showing both the layered structure and crossbar connections of one TFT. (**d**) Cross-sectional schematic of a single printed TFT with detailed material annotations. (**e**) Output characteristics of a single representative TFT. (**f**) Subthreshold and gate leakage characteristics of all TFTs in a 2×2 pixel array with the 100% device yield that is required for a functional array. Dashed line represents a subthreshold swing of 120 mV/dec.

stop the etch, strip the resist, and clean the substrates.

All-carbon recyclable TFTs were aerosol jet printed on the patterned ITO substrates according to the methods established in our previous reports [18], [35]. Briefly, source/drain electrodes were printed first with a water-based graphene ink (MilliporeSigma, 808261-10ML) diluted to 33 mg/mL. The channel was printed next with a toluene-based CNT ink (NanoIntegris, IsoSol-S100) diluted to 10 µg/mL. Since this ink is composed of CNTs that are kept in suspension by wrapping polymers, printing was followed by a 10 min soak in toluene at 80 °C to remove excess polymer. The crossbar spacer was then printed from a water-based CNC ink (Cellulose Lab, CNC-Slurry-HS) diluted to 60 mg/mL. The gate dielectric was printed with the same CNC ink but with 0.5 mg/mL NaCl added to increase the ionic content and capacitance of the gate dielectric. Lastly, the top gate and crossbar bridge were printed with the same graphene ink previously used. A complete TFT is shown in Fig. 1b-d.

After printing, the water-recyclable transistors were waterproofed by spin coating with poly(methyl methacrylate) (950k PMMA A5), followed by a soft bake at 180 °C for 3 min. Substrates were allowed to cool in air for 1 hour to allow time for the PMMA and CNC dielectrics to rehydrate and stabilize in the ambient environment. Next, the PMMA passivation layer was patterned: using a 3D-printed shadow mask, the resist was exposed to a 254 nm lamp (Analytik Jena, UVS-26) for 16 hours and developed in a 3:1 IPA/H_2O mixture for 5 min. This removed PMMA over the transparent pixel electrodes to enable the use of a variety of dielectrics for the alignment layer in the LCD pixel stack.

Finally, after device passivation, the LCD pixel stack was constructed. The substrate received a final spin coating of the alignment layer, which was either a more dilute PMMA (950k PMMA A2) or a more dilute CNC (10 mg/mL). A second ITO-coated glass substrate was coated with an identical alignment layer, both substrates were mechanically polished at 90-degrees to each other, and finally they were epoxy bonded with ~10 µL of nematic liquid crystal 5CB (MilliporeSigma, 328510-1G) sandwiched between to form the complete LCD.

III. Results and Discussion

The TFT performance characteristics are shown in Fig. 1e-f. Despite the thick, printed gate dielectric and long channel, the devices reach saturation (Fig. 1e) at low drain-source voltage (V_{DS}), and a similarly low gate-source voltage (V_{GS}) modulates the channel (Fig. 1f). The low subthreshold swing (SS), low V_{GS} needed for switching the TFTs, and corresponding low V_{DS} needed to reach saturation, are all a result of the strong capacitance of the electric double layer that forms inside the CNC ionic gate dielectric, immediately at the interface to the CNT thin-film channel. This was characterized in detail in prior work [35], which showed that the addition of NaCl to CNC dielectrics lent them ionic behavior very similar to the widely studied EMIM-TFSI copolymer ion gels.

For this application, the TFTs are connected in a crossbar matrix structure, where TFTs in each row have connected gates and TFTs in each column have connected drains (Fig. 1a). Therefore, it was necessary for the TFTs to be printed with an integrated spacer and bridge to allow the drain line to pass over the gate line (see Fig. 1b-d). However, this overlap forms a parasitic capacitance between the gate and drain, and any leakage through the spacer would contribute to the gate current (Fig. 1f) and could degrade the subthreshold swing. So, unlike in the CNC gate dielectric where high capacitance was desirable, NaCl was not incorporated in the spacer dielectric to minimize parasitic capacitance and leakage.

In addition to parasitic capacitance, the crossbar structure also introduces series resistance. With an aspect ratio (length/width) of 150 and a nominal sheet resistance of 15 Ω/sq, the ITO crossbars were designed to contribute only ~2 kΩ of resistance from end-to-end, and this was confirmed by electrical measurements. However, the resistance of printed graphene crossing over a printed CNC spacer was more variable than anticipated, contributing an additional 5-25 kΩ per bridge (i.e., up to ~50 kΩ to the total drain-line resistance, dropping 50 mV per µA of operating current). While this did not prevent the realization of functional LCDs, future work should explore reducing bridge resistance.

As previously noted, the as-printed TFTs are water-soluble, which enables reclaiming the constituent inks through a water-activated recycling process [18], as illustrated in Fig. 2a. This means it would be necessary to use all non-aqueous processing in the LCD layers on top of the TFTs – in particular, it would prevent the use of water-based recyclable CNC as the LCD alignment layer. Hence, we developed an encapsulation

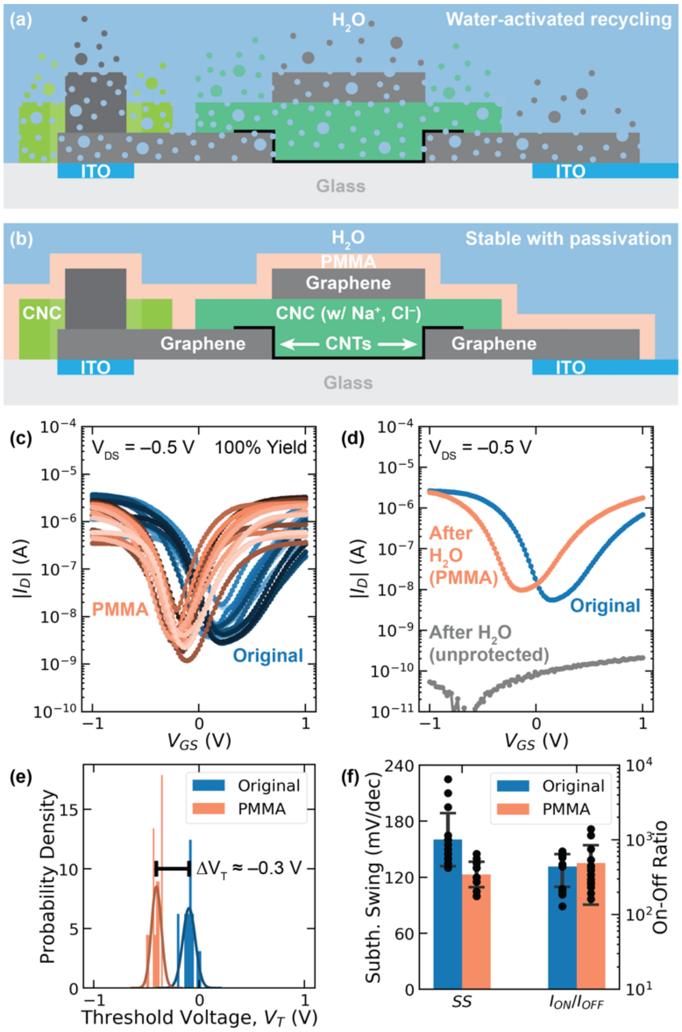

Fig. 2. Waterproofing recyclable transistors that otherwise dissolve in water. (**a**) Cross-sectional schematic demonstrating water solubility of all-carbon recyclable TFTs. (**b**) Cross-sectional schematic highlighting the use of targeted PMMA passivation for protecting printed TFTs. (**c**) Subthreshold characteristics of fourteen devices before and after PMMA passivation, showing 100% yield and a slight threshold voltage shift. (**d**) Subthreshold curves of a representative device before and after spin coating in H_2O (with and without passivation), demonstrating how the PMMA layer addresses an application-specific need for compatibility with aqueous processing in LCD fabrication. (**e**) Histograms and Gaussian probability density of threshold voltage before and after PMMA passivation, showing an average shift of -0.3 V. (**f**) Bar graphs of subthreshold swing and on/off-current ratio before and after PMMA passivation. Error bars show one standard deviation.

strategy using PMMA (another carbon-based insulator), as shown in Fig. 2b. The TFTs were coated in a nominally 400 nm thick film, and this passivation layer was confined to the device regions by using ultraviolet lithography to pattern the film. This process is simple and minimally disruptive to the TFTs with 100% fabrication yield (Fig. 2c), also causing only a moderate and temporary threshold voltage (V_T) shift that reverted after 24 hr. Afterwards, the TFT backplanes are compatible with aqueous deposition of the LCD layers (see Fig. 2d). Distributions of V_T (Fig. 2e), as well as SS and on/off-current ratio (Fig. 2f) show a benign impact of this passivation on key device metrics, including a -0.3 V shift in V_T and 37.5 mV/dec decrease (or 23% improvement) in mean

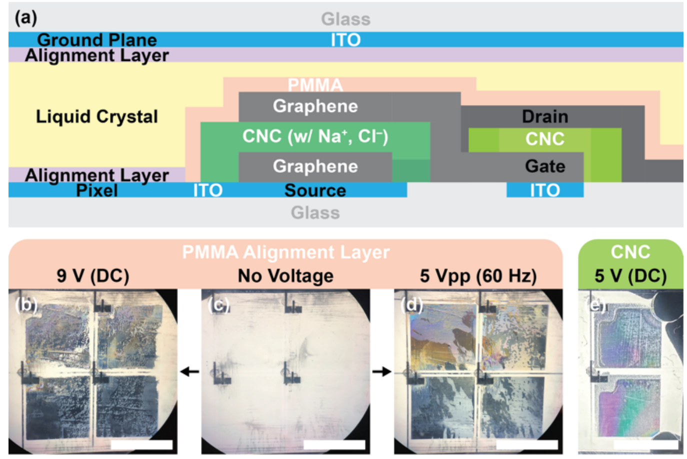

Fig 3. Characterization of LCDs driven by fully printed, recyclable TFT backplanes. (**a**) Cross-sectional schematic of final full LCD stack. (**b**) Photo of LCD array switched on with 9 V applied to gate and drain lines relative to the ground plane. (**c**) Photo of LCD array switched off (all inputs grounded). (**d**) Photo of LCD array with 60 Hz, 5 V peak-to-peak signal applied to gate and drain lines. (**e**) Photo of dual-pixel LCD array with recyclable CNC used as the alignment layer and operating at 5 V. In all photos, arrays are held between two perpendicularly-oriented polarization filters, which enables the liquid crystal to block light under an applied voltage. All scale bars are 1 cm.

SS. Note that the common TFT metric of field-effect mobility was not extracted here as the frequency-dependent capacitance of the CNC can lead to exaggerated reporting of that metric.

The final LCD cross section is shown in Fig. 3a. Two different alignment layers were fabricated: a 2×2 array with PMMA as the alignment layer (Fig. 3b-d), and a 2×1 array with CNC as the alignment layer (Fig. 3e). In both cases, the target alignment layer thickness was <100 nm (thinner than the waterproofing passivation layer) to minimize the required operating voltage. Still, while the TFTs can operate at lower voltages, the array required at least ~5 V to switch the liquid crystal, consistent with similar reports [25]. For simplicity, due to the discrepancy between these operating voltages (5-10 V) and the maximum safe steady-state gate-to-drain or gate-to-pixel voltage for the TFTs (<2 V), the entire array was turned on and off together to avoid damage to the backplane. Promisingly, the array had no issue operating in an alternating current (AC) mode at 60 Hz (Fig. 3d), which as expected also produced superior pixel edge fidelity compared to direct current (DC) operation (Fig. 3b). With more precise voltage tuning or sophisticated pulsed operation it would be possible to achieve single-pixel control.

IV. Conclusion

In conclusion, we report the first demonstration of an LCD driven by an all-carbon recyclable printed transistor backplane. This was achieved through integration of the printed TFTs with a transparent ITO crossbar structure, along with the development of a device passivation strategy that enabled the use of aqueous recyclable materials not just in the backplane but also as the liquid crystal alignment layer. This work shows the promise of bringing an emerging recyclable device technology to displays with the goal of more environmentally friendly electronics.

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