Organic Thin-Film Transistor Formulations Proven in Mass Production

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Abstract

We demonstrate the development and optimization of a full stack of organic thin-film transistor (OTFT) materials, which has now been proven in mass-production of a consumer electronics device. New materials and formulations enable improved compatibility with typical flat panel display (FPD) line manufacturing tools and processes, while maintaining transistor performance above that of amorphous silicon (a-Si).

Author Keywords

Thin-film transistor; OTFT; EPD; Organic dielectric; Organic semiconductor; Integration; Adhesion

1. Introduction

Organic thin-film transistors have been of interest to the electronics industry, and in particular displays, for at least 20 years [1], due to the promise of low temperature manufacturing on a variety of flexible and stretchable plastic substrates, enabling display form-factors that are not achievable with inorganic transistors.

In the early years, the focus was on improvements in charge-carrier mobility of the organic semiconductor (OSC), often at the cost of processability and robustness of the materials stack. The critical breakthrough was the development of air-stable polymer semiconductors with mobility exceeding that of a-Si, which was regarded as a key display industry benchmark. [2] The move from crystalline small-molecule OSCs to polymers produced two improvements, which have been essential to the state of the art.

The first is that an all-polymer stack is intrinsically stretchable, giving greater design-freedom compared to, for example, the LTPS transistors most used in flexible OLED smartphone displays. Inorganic ceramics, like LTPS, can crack under the strain of bending so must be placed as close as possible to the neutral axis, whereas organic polymers have no such constraints.

The second is that the move from highly soluble small molecules to engineered polymers allowed for the removal of highly fluorinated dielectrics and solvents from the stack, greatly widening the space for development and optimization of the passive layers in the stack [3] and sidestepping the regulatory and environmental concerns around the use of fluoropolymers.

FlexEnable has been developing world-leading capability in organic TFT processes and stacks for many years at this point and, since the acquisition of Merck KGaA's organic TFT materials portfolio in 2019, has added a significant focus on fundamental materials and formulation development to this capability.

The outcome of this work, following a concentrated development phase with manufacturing partners Giantplus (Taiwan) and DKE (China), is the successful mass production of organic TFT backplanes and integration into the first consumer electronics device – the Ledger Stax crypto wallet – with an EPD display curved around 180° exterior bend with a 3mm radius as shown in Figure 1. [4] The development of ever more resilient and processable organic-TFT formulations is essential to the continued development of new display form-factors and applications.

Figure 1 The Ledger Stax product showing the OTF/EPD display wrapped around the full 180° with a radius of 3 mm.

In this paper, we report on new dielectric formulations designed to improve the adhesion of the critical layers in the stack during exposure to common FPD manufacturing processes. Adhesion between the stretchable polymer layers is critical to stack integrity as the backplane is curved whilst also allowing a wider process window in production. We also report on the organic-TFT performance using the new formulations, as reliable operation is essential for the long-term viability of any consumer electronics product.

2. Experimental

The results presented in this paper were obtained using FlexEnable's 14" prototyping line and the FlexiOM organic TFT material set. The simplified top-gate materials stack used is shown in Figure 2 and is analogous to the stack used in production of the Ledger Stax.

The flexible substrate was a 125 µm PEN film, laminated to a carrier glass with a double-sided adhesive to allow the substrate to be handled by standard FPD manufacturing tools. All the process steps utilize equipment commonly found in fabs: DC sputtering for metals, and standard photolithography including acid etchants for patterning steps. Since the semiconductors and dielectrics are solution-based and are deposited by spin or slot-die coating, no CVD tools are required. All process steps are also carried out at below 100° C, saving energy vs typical inorganic TFT processing at 350° C or more.

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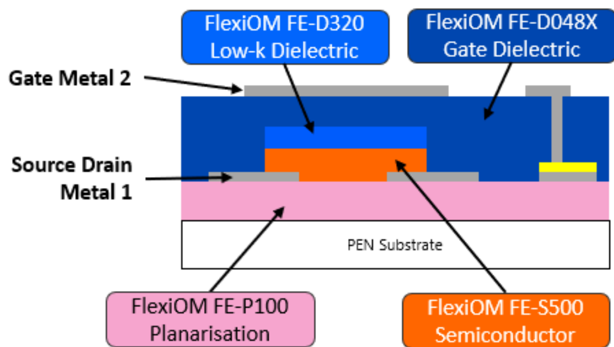


Figure 2 The top-gate FlexiOM materials stack used in this paper. The full EPD stack contains further screening and pixel dielectric layers, but this stack is sufficient to test the adhesion and TFT performance.

The first layer deposited was FlexiOM FE-P100, a new 365 nm UV-cured planarization that provides a consistent surface for adhesion and TFT processing independent of the choice of substrate. Typical thickness for the P100 layer is 1 – 3 µm, depending on the substrate choice and design requirements of the stack. In this case, a 1.2 µm layer was spin-coated, baked dry and then cured with a UV dose of 4 J/cm^2 and hard baked at 85° C for 5 minutes on a hot-plate.

A 40 nm layer of silver alloy was sputtered on top of the base-layer and patterned using an FPD photoresist and phosphoric-acetic-nitric acid (PAN) etchant to form the source and drain electrodes. The patterned electrodes were treated with a charge injection SAM to improve TFT performance and uniformity, and the FlexiOM FE-S500 polymer OSC was spin-coated to produce a film of approximately 50 nm thickness. Immediately after that, the first low-k dielectric layer, FlexiOM FE-D320 was spin coated, to produce a clean and dipole-free interface for efficient charge-transport.

The OSC and first dielectric layer were patterned together using a standard FPD photoresist and 365 nm mask exposure tool. The photoresist was developed using 2.38% TMAH in water, and dry-etched using an O_2/CF_4 reactive ion etch (RIE).

Following photoresist strip, the second dielectric layer was spin-coated. The current product, FlexiOM FE-D048X, was compared to the newly developed FE-D048XR. FE-D048XR contains novel adhesion additives, developed and synthesized in house and intended to improve adhesion to a variety of underlying layers in the patterned stack. After spin-coating and drying, both dielectrics were UV cured using a 365 nm LED lamp, resulting in a total dielectric thickness of ~750 nm. This double-layer dielectric process allows for good isolation of the OSC within the TFT channel, whilst protecting it from subsequent metal sputtering and etching processes.

Contact Vias were then dry-etched to provide electrical connections to the source-drain metal layer, before the molybdenum alloy gate metal was sputtered and etched by standard photolithography.

Electrical testing was performed using an automated Agilent 4155C parameter analyzer on standardized transistor test substrates contacted via a zero-insertion-force connector socket.

Adhesion testing was done at various stages through the process using a Mecmesin Imperial 1000 90° peel-force tester with 25 mm wide TESA 4965 tape. Substrates were prepared as shown in Figure 3. With the test piece securely fastened to the test bed, an initiation cut was made at the start of the peel using a scalpel, and the tape was firmly applied to the surface before being attached to the peeling fixture. Cuts were then made along the sides of the tape to ensure that only the area of the tape was peeled. Adhesion values were taken in units of Newtons per mm of tape along the peel face.

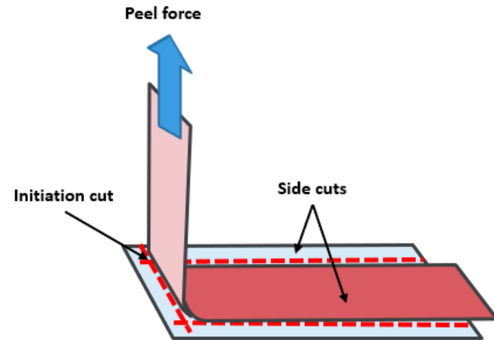


Figure 3 Substrate preparation for 90° peel test. An initiation cut is made near the edge of the substrate, and Tesa 4965 tape is firmly applied to the surface. Relief cuts are made along the sides of the tape.

Anomalous peaks in adhesion force can often be seen at the start of the peel, although the initiation cut should minimize this, and these can be discarded, with results taken from the stable region of the peel. Adhesion is quoted per centimetre of tape at the peel interface.

3. Results and Discussion

Adhesion of FE-P100 to substrates: Figure 4 shows the adhesion of FE-P100 films spin-coated on a range of substrate materials. In addition to the PEN film used in the Ledger Stax process, adhesion was also tested on tri-acetyl cellulose (TAC) – FlexEnable's standard substrate for optical applications – and display glass, which is routinely used in lab-scale formulation testing.

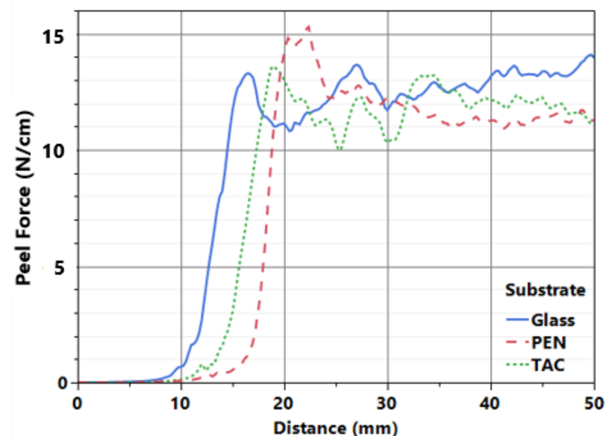
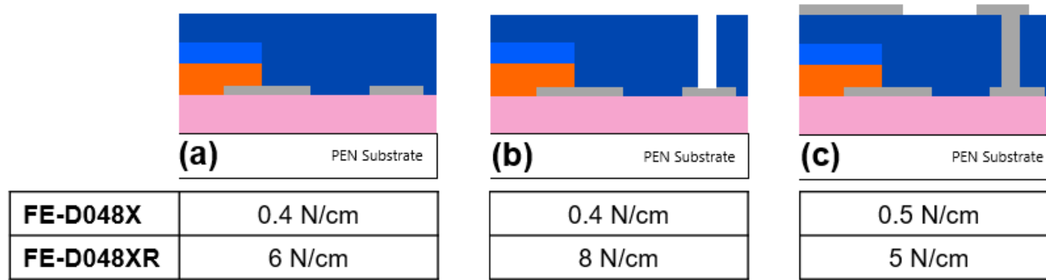


Figure 4 90° peel test results for FE-P100 on a range of substrates. In this case, the tape peeled from the surface of the film, allowing us to infer the underlying adhesion to be greater than 10 N/cm.



	(a)	(b)	(c)
FE-D048X	0.4 N/cm	0.4 N/cm	0.5 N/cm
FE-D048XR	6 N/cm	8 N/cm	5 N/cm

Figure 5 Adhesion of D048XR vs D048X as the film was taken through the standard backplane production process. Adhesion is at least an order of magnitude higher for D048XR after (a) film coating and UV cure, (b) photolithography and dry-etch of contact via and (c) gate metal sputter, photolithography and acid etch.

After the slack in the tape was taken up at around 10 mm peel length, in all cases, the tape peeled from the surface of the FE-P100. Since adhesion of the FE-P100 to the substrate is maintained, we can infer that the adhesion of FE-P100 is greater than 10 N/cm independent of substrate choice.

D048XR Adhesion During Processing: Adhesion of the FE-D048XR product compared to the previous generation material is shown in Figure 5. In this trial, the full TFT display backplane process was run, with adhesion tests carried out after key process steps as shown in the stack cross-sections. The adhesion of the D048XR film after curing was 6 N/cm (a), which is more than 10 times greater than D048X.

After photolithography and RIE patterning of the contact vias (Figure 5b) the adhesion was tested again, giving 8 N/cm for D048XR. At this point, the film has been exposed to the solvent in the photoresist, TMAH developer, stripper and the moderate vacuum of the RIE chamber.

The final adhesion test was carried out after the sputtering and patterning of the gate (Figure 5c), adding exposure to DC sputtering and PAN etchant. Adhesion of D048XR at this point was 5 N/cm and still an order of magnitude higher than D048X.

The process steps outlined above comprise a representative sample of those the material is exposed to in the full manufacturing process and, whilst we do see some variation in adhesion between tests, the trend of increased adhesion for D048XR is maintained, confirming that the adhesion is stable in a wide range of conditions.

Transistor Performance: Following the adhesion tests, the electrical performance of organic-TFTs produced in the same process was measured. Figure 6 shows the transistor transfer scans for D048X (blue) vs D048XR (red) with channel length L = 5 μm and channel width W = 20 μm, with a gate voltage (V_G) range of -20 V to 10 V and drain voltage (V_D) = -1 V in the linear regime (a) and V_D = -20 V in the saturation regime (b). For each material, six transistors each on two substrates were measured, and the mean values plotted. This results in transfer curves, which clearly represent the performance differences between the materials.

The transistor switching performance remains largely unchanged between D048X and D048XR. Off current (I_{OFF}) is approximately 5 x 10^{-12} A for both materials in either the liner or saturation regime. Turn-on voltage (V_{ON}), calculated as the V_G at which the drain current passes 1 x 10^{-9} A, is shifted negative by 0.8 V for D048XR, but this shift does not present any issues for driving EPD or LCD displays.

If the adhesion additives introduced any charged or chemically reactive species into the TFT channel, we would expect to see it here. The small difference in V_{ON} suggests that there may be some fixed charge in the D048XR dielectric contributing to the shift in effective gate voltage of the TFT, but otherwise the performance is equivalent to D048X.

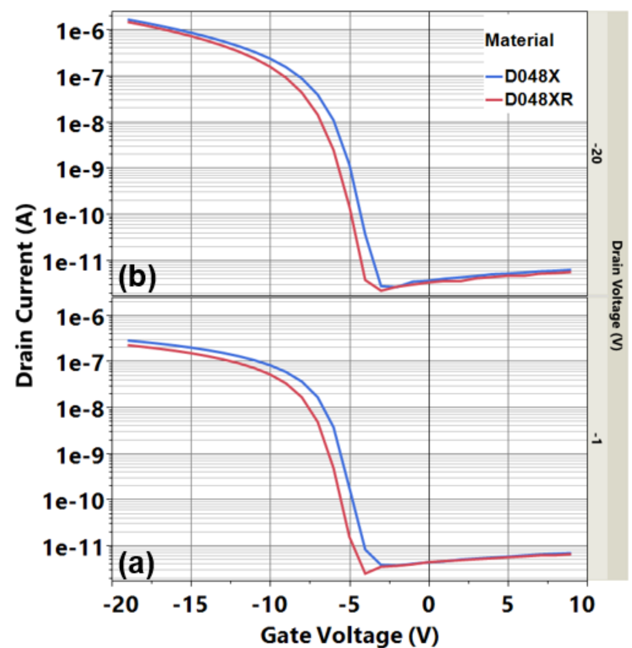


Figure 6 TFT transfer scans for D048X (blue line) and D048XR (red line) averaged for 12 TFTs each with L = 5 μm, W = 20 μm. Transfer scans were taken with V_D = -1 V (a) and V_D = -20 V (b), for V_G = 10 V to -20 V. Performance is unchanged, apart from a 0.8 V negative shift in V_{ON}.

4. Conclusions

Based on the adhesion improvements reported here, the robustness of the FlexiOM organic-TFT stack has been greatly improved. A truly substrate-independent planarization layer, FE-P100, allows for application-specific substrate selection without compromising compatibility with the TFT stack, whilst a factor-of-ten improvement in adhesion of D048XR has been achieved with minimal impact on critical TFT performance metrics.

The improved adhesion also appears to be resilient to a wide range of standard chemical processes, which can be challenging for

organic transistor materials where electrical performance is usually the primary design factor. Improved adhesion through the organic TFT stack also places fewer constraints on both photo-mask design and display bend at final device integration, so the benefits should be felt throughout the production chain.

5. Impact of Your Research

We expect these improvements in the strength of the stack to broaden the process window for manufacturing and hasten the implementation of organic-TFT materials into more fabs and, ultimately more products.

The addition of FE-P100 to the FlexiOM materials portfolio also gives us greater power to adjust the properties of the whole stack through further chemistry or formulation modifications, targeting a wider range of applications in the transmissive space, where e.g. optical properties are more critical.

6. References

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