

Large-Scale Manufacturing of Polymer Planarization Layers

Polymeric planarization layers play a pivotal role in OLED displays' thin-film encapsulation. With its low material consumption, deposition geometry flexibility, high throughput, and reliability, inkjet printing is the preferred deposition technology for such films.

by Lorenza Moro and Christopher R. Hauf

FLEXIBLE OLED DISPLAYS HAVE BECOME A MAINSTREAM technology in high-end mobile consumer applications. The first commercial active-matrix OLED (AMOLED) display on plastic foil using thin-film encapsulation (TFE) with a curved appearance was sold in limited numbers in 2013. Currently, the flexible OLED market represents about half the OLED mobile devices market—that is, 240 million units, which is roughly 16 percent of the total almost 1,400 million units shipped in 2020.¹ The flexible OLED market share expands each year, with it expected to double to 30 percent in 2025. While Samsung Display Corporation (SDC) is still the major supplier (90 percent), massive investments (mainly in China) suggest that flexible displays' price will decrease in the next few years, and this technology will become dominant for mobile displays.²

AMOLED displays' large-scale mass production deployment started when TFE made new functionalities possible for commercial deployment by allowing flexible displays' fabrication on plastic foils with a total thickness of a couple hundred microns. Thinner displays help reduce the final mobile devices' thickness and weight, making possible high-end, larger-size phones sold at premium prices. In the following generations, the substrate's flexibility has allowed bending back the portion of the display that supports connections to external contacts. "All Screen," "Infinite Display," "Near Bezel-less," and "Waterfall" displays have been deployed in stylish handsets where many of the accessory components (such as cameras, buttons, and speakers) are integrated in the displays area itself. Royole, Samsung, Huawei, and Motorola put fully foldable displays for foldable

phones on the market in 2019. This format is gaining popularity, leading to greater adoption with 2020's improved models and plans for 2021.³

OLED displays' commercial deployment requires high-yield and robust encapsulation technology because OLED materials and devices are extremely sensitive to moisture and oxygen contamination. In addition, encapsulation is a back-end process, so any failure affects the final yield. The possibility of flexible OLED displays on plastic foil and encapsulated with thin films had been proposed early in the history of OLED devices, but the debate on its commercial feasibility was quite animated and long-lasting. This is because of the extremely demanding requirement to TFE: barriers with water vapor transmission rates (WVTR) lower than 10^{-6} g/(m²/day) to avoid short-term degradation of devices by black spot formation; oxygen transmission rates (OTRs) lower than 10^{-4} cm³/(m² day) to prevent middle- or long-term degradation of light emission efficiency by oxygen; and no point defects. Usually point defects are created by particles introduced by the OLED active layers' evaporation process with fine metal masks (FMMs) used in strict proximity (~10 micrometers, or μ m) to the display glass. Advanced tools and strict procedures mitigate—but do not avoid—the display glass' particle contamination even in mature production lines.

Inorganic layers are an effective barrier to moisture and oxygen diffusion. The limitations imposed on the deposition process by OLED materials' inherent sensitivity to temperature ($T < 80 - 100^{\circ}\text{C}$) and plasma processing has made the multilayer (ML) approach—based on a three-layer stack of inor-

ganic, polymer, and inorganic (**Fig. 1**)—successful for AMOLED's mass-production encapsulation.

Vitex⁴ proposed the ML-TFE technology to the industry in the early 2000s; the company demonstrated successful encapsulation by 1 to 2 dyads, making the technology interesting for commercial applications.⁵ With further development, Samsung deployed the technology at large-scale with the introduction of flexible displays in the Galaxy Edge series, with a curved display at one or two sides. In following generations, Samsung introduced direct fabrication of the touch screen on the TFE structure, thereby further reducing the displays' thickness and pushed forward the integration of functions in the display area by introducing in it holes to allow the assembly of cameras and sensors under the display itself. Finally, TFE encapsulation has been optimized for fully foldable displays for foldable phones by Samsung and others.

Recently, the same 1.5 dyads TFE structure plasma-enhanced chemical vapor deposition (PECVD) silicon nitride (SiN_x)/ink-jetted-polymer/PECVD-SiN_x has been adopted by most of the new flexible OLED display suppliers in China, Taiwan, and Japan. The same structure is implemented in the OLED display fabricated by Samsung, deployed by Apple in some iPhone models released after 2017.

TFE's Planarization Layers

The need for a ML structure with a polymer layer sandwiched between two inorganic layers is well-established and accepted among experts in the fields of ultra-barrier technology.

A first important function of the organic layer in a barrier structure is providing a smooth surface that allows optimum nucleation conditions to inorganic layers deposited at a low temperature. Inorganic films deposited on rougher surfaces grow with columnar grain structure and lower density, while the film grows dense and amorphous on the smooth surface.⁶ Polymer layers of suitable thickness also can cover the morphological structure present on the surface of the display to be encapsulated. Morphological features may be part of the display itself—such as pixel defining layers (PDL) or post separators for mask (PS)—or particles deposited on the surface by previous fabrication steps, such as fine metal masks (FMM) masks for evaporating red, green, and blue (RGB) pixels.

A second important function of polymer layers is decoupling the inorganic layers that constitute the real barrier to permeation. In this configuration, vapor and gas molecules—notably H₂O and O₂—enter the ML barrier through a defect of the more external inorganic layer and laterally diffuse through the polymer film to reach a defect in next inorganic layer, thereby permeating to the OLED active layers via a tortuous path. Theoretical modeling and experiments have demonstrated that vapor permeation through multilayer ultra-barrier structures is not described by steady-state diffusion. Ultra-barrier performance is the effect of exceptionally long lag times created by an extremely long effective path for gas diffusion.⁷ Finally, polymeric layers in a barrier structure improve its mechanical reliability and flexibility. Thin inorganic layers as the ones used in



Fig. 1.

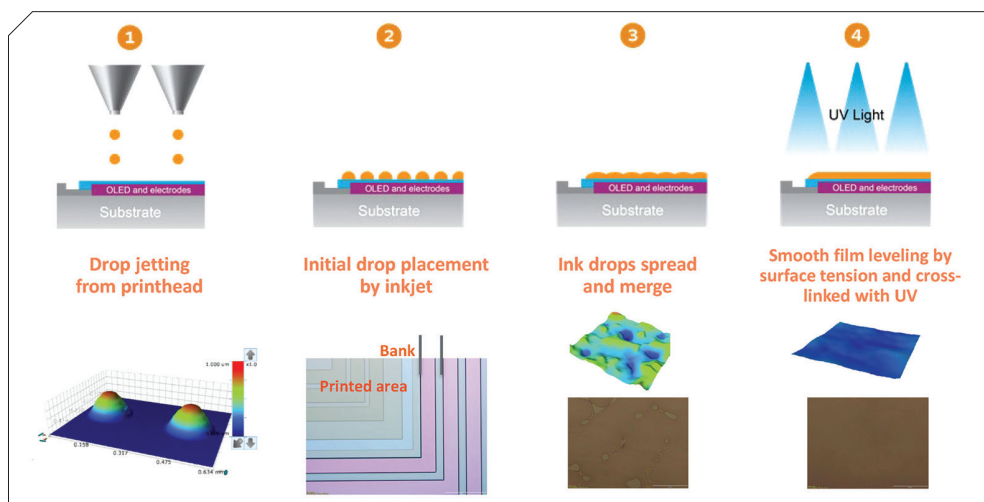
Typical multilayer thin-film encapsulation (ML TFE) structure used in current smartphone displays. SiN_x = silicon nitride.

an ML barrier are intrinsically more flexible than a thick inorganic film. In addition, less fragmentation and cracking are possible for brittle ceramic layers sandwiched by compliant organic layers.

Some researchers also have claimed an active role for the polymer used in multilayer barrier structures. Defect sealing by filling them with a gas phase or liquid polymeric precursor has been credited with improving barrier performance.⁸ This “repair” mechanism may be relevant in reducing permeation in inorganic films of limited barrier performance, but it is not enough in transforming them in ultra-barriers, and it compromises mechanical and optical film properties.

Researchers have proposed several methods for the deposition of an organic layer either based on plasma polymerization, resulting in conformal coating or deposition of liquid formulations, followed by polymerization in general induced by ultraviolet (UV) exposure:

- Parylene⁹ and initiated chemical vapor deposition (i-CVD)¹⁰ are vacuum thermal deposition techniques of polymers prosed in academic literature for fabricating multilayer barriers, but have never been used in mass manufacturing for intrinsic limitations and lack of commercially available reactors.
- Several groups have proposed organic-like soft films by plasma polymerization since TFE's early onset.¹¹ One implementation¹² uses Applied Material PECVD systems—the tools with the largest installation footprint for TFE inorganic layer deposition in the flexible OLED industry. The authors propose a flowable HDMSO film as planarization layer in a three-layer silicon nitride (SiN_x)/hexamethyldisiloxane (HMDSO)/SiN_x structure for flexible applications. The full TFE structure's deposition in a unique cluster tool is presented as a unique advantage. However, the structure seems to have had a lim-

**Fig. 2.**

Critical steps in forming film by inkjet printing (IJP) deposition: 1-jetting; 2-drop placement; 3-film formation; 4-curing. UV stands for ultraviolet.

VTE process' advantage is the high deposition rate with high polymer film quality and good uniformity. Challenges in hardware (uniformity and chamber contamination) and material utilization made the process difficult to scale up in early studies and proposals of using inkjet printing polymers for TFE application in mass

ited commercial success in mobile applications. Two main drawbacks of such an approach are the limited planarization effectiveness and the need of patterning masks that are expensive for their intrinsic cost and the required cleaning cycles connected with their use in mass manufacturing limiting the throughput. A narrower display edge width may be an advantage over planarization by liquid deposition, where the width is limited by fluid rheology. However, this advantage is watered down by the inability or extreme complexity of realizing features such as notches or camera-on-display that have become common form factors in current mobile devices. This planarization approach may become suitable for displays fabricated with intrinsically cleaner processes and requiring a narrow bezel, such as microdisplays.

- Planarization layers fabricated by liquid precursors' deposition, followed by UV polymerization were early implemented with success in TFE and have become the dominant planarization approach. In the early implementation screen printing,¹³ thermal evaporation,¹⁴ and inkjet printing processes have been used.^{15,16} Screen printing is a cost-effective method of making patterned thick film up to 100 μm , but coating of thin films ($<10 \mu\text{m}$) is difficult. Screen deformation makes it hard to control film edge to a narrow range, resulting in a wide display bezel outside the active display area. These undesired features have made screen printing unsuitable for mobile display panels' TFE; but they are compatible with fabricating planarization layers with a function other than TFE in OLED TV applications.
- Vacuum thermal evaporation (VTE) is a process that has demonstrated mass production capability in making ultrasmooth and pinhole-free polymer film on continuous meter-wide web roll. Vitex Systems adapted it for polymer films' deposition on discrete substrates. The method was used widely in R&D tools for TFE of rigid and flexible OLEDs on 200-mm substrates and successfully scaled to 1/4 G5.5 for flexible OLED mass production.¹⁷ The monomer vapor produced by an atomized/sprayed liquid into an evaporator diffuses to a vacuum chamber, where it condenses on the shadow-masked substrate as a liquid film and subsequently is cross-linked to form a polymer film by exposure to a UV source. The vacuum-compatible

manufacturing.¹⁶

- Inkjet printing (IJP) allows direct patterning of polymer areas with uniform thickness, high precision of location, and definition of edges—at high speed and with efficient use of material. It is therefore an ideal coating method for TFE planarization layers' deposition in mass production. **Fig. 2** schematizes the critical steps required to achieve production-worth planarization layers: Step 1, drops of known volume are ejected by a piezoelectric printhead at frequencies that can be greater than 20 kilohertz (kHz); Step 2 they are placed on the substrate at a precise location by coordinating drop ejection with accurate control of printhead and substrate movements; Step 3 drops spread and merge for a pre-select leveling time to form a continuous liquid film (note that proper tool design and configuration may minimize the impact of leveling time on throughput); Step 4 the smooth, leveled liquid film is polymerized by UV radiation. Currently, solid-state LED light with a narrow wavelength distribution around 395-nanometers (nm) are used as sources. Following the ejection of ink droplets by the printhead nozzles, a few factors affect the polymer film's formation on the substrate surface—including ink and substrate wettability, ink viscosity, surface and ink contamination, airflow, substrate roughness, and even electrostatic charge on the substrate.

IJP Planarization Layers in Mass Production

IJP intrinsically meets requirements for being the technology of choice in coating planarization layers of TFE structures. A few more challenges had to be overcome to use and upscale IJP coating of the organic layer in TFE, to make it manufacturable in masses.

Kateeva is now the leading supplier of TFE IJP tools to the OLED industry, with a substantial share of the market in Korea, China, and Taiwan. In typical OLED production lines, the IJ printer is nestled between two PECVD clusters at the end of the N2-enclosed OLED fabrication line (**Fig. 3**). **Fig. 3's** configuration meets a takt time of less of 60 seconds. Kateeva's product portfolio for TFE planarization layers covers the full

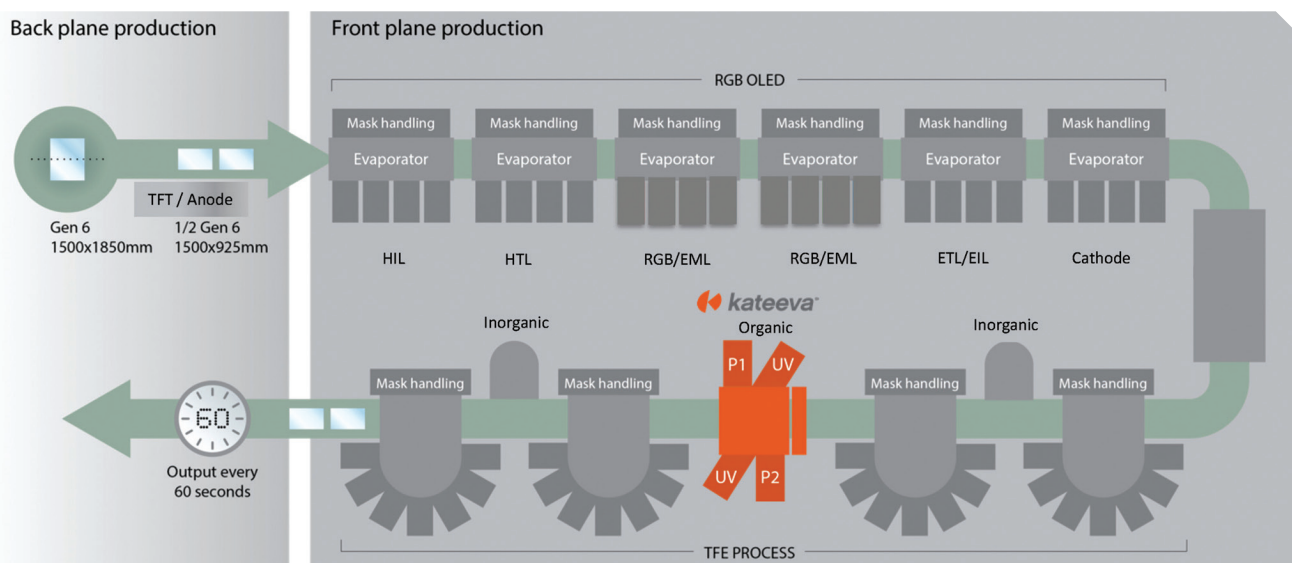


Fig. 3.

Schematic of mass producing the OLED deposition and encapsulation line for flexible displays. EIL = Electron Injecting Layer; EML = Electron Emitting Layer; HIL = Hole Injection Layer; HTL = Hole Transport Layer; P1/P2 = Printer 1/2; and RGB = red, green, and blue.

display development trajectory and its increasing mother-glass size. Having consistency among the tools' hardware and software allows for an easy transfer and upscale of processes and seamless testing and qualifications of new materials. **Fig. 4** shows a YIELDjet Flex 750 (650 × 750 mm) installed at Kateeva's headquarters for TFE demos and internal development.

One important differentiation in the high-end mass production tools is that coating occurs in an inert atmosphere. This is required because, at this process step, OLED devices are protected only by an inorganic layer prone to defects that could evolve quickly into blackspots on the OLED device by moisture and oxygen permeation. Additionally, any moisture and contamination from the environment could be trapped in the liquid or cross-linked organic film during processing (jetting, leveling, and curing). Such impurities trapped inside the barrier after the subsequent deposition of the inorganic layer(s) would slowly diffuse to the OLED through pinholes and degrade them. Optimizing the enclosure volume, N₂ flow, and overall design have been considered to minimize particle addition and decrease recovery time after maintenance.

Long- and short-range film thickness uniformity is an important parameter in TFE encapsulation of top emission displays because it may lead to a luminance non-uniformity of the light that the display emits (or reflects). In this context, the thickness non-uniformity is named mura from the Japanese word meaning unevenness or irregularity. In IJP printing, mura can be caused by using a regular grid of ink drop patterns that reproduce nozzle-to-nozzle variations in the print heads. Mitigation of printing mura can be done by a statistical approach using randomization in nozzle firing or by using more sophisticated algorithms that use multiple sources of data to accurately and precisely deposit the layer.

A different cause of thickness non-uniformity is chuck mura, produced by the interaction between the chuck and substrate. Thermal gradients affecting the spread and merge of drops are created by contact between the substrate and chuck. Uncontrolled variations of the distance between the printhead and substrate can affect the droplet landing position and, therefore, the distribution of the ink on the substrate. High-end mass production printers employ proprietary floating-glass technology that, by using pressure and vacuum holes, prevents contact and precisely controls the relative printer-head-substrate position. Non-compensated electrostatic charges may also create defects in the liquid ink distribution, so ionizers able to compensate tribological charges should be used.

High reliability and printer speed are necessary to meet throughput and uptime process requirements. The printheads used to apply the organic material are based on piezo technology. When a voltage is applied to the piezo head, it triggers actuators that jet out ink through nozzles in precisely measured droplets with great speed and accuracy. With this technology, droplets with from few to tens of picoliters can be ejected at frequencies up to a few tens of kHz. Industrial-grade piezo printheads have a lifetime of billions of drop ejections and could last many months in a 24/7 production environment depending on several factors. Multiple printheads on each printer, along with integrated system design, allow the total system to meet 60-second takt time for one sheet of G6H-sized glass (925 × 1,500 mm).

The inkjet systems are flexible in accommodating multiple dis-

Fig. 4.

YIELDjet Flex 750 installed at Kateeva's headquarters for TFE demos and internal development.



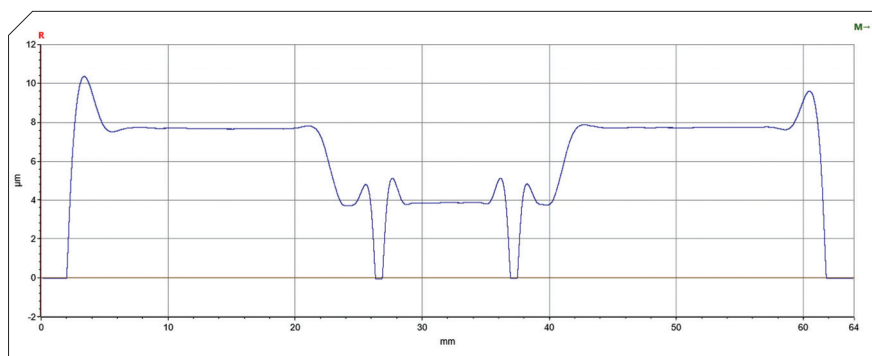


Fig. 5.

Profile of the multithickness planarization layer deposited by IJP with no edge compensation.

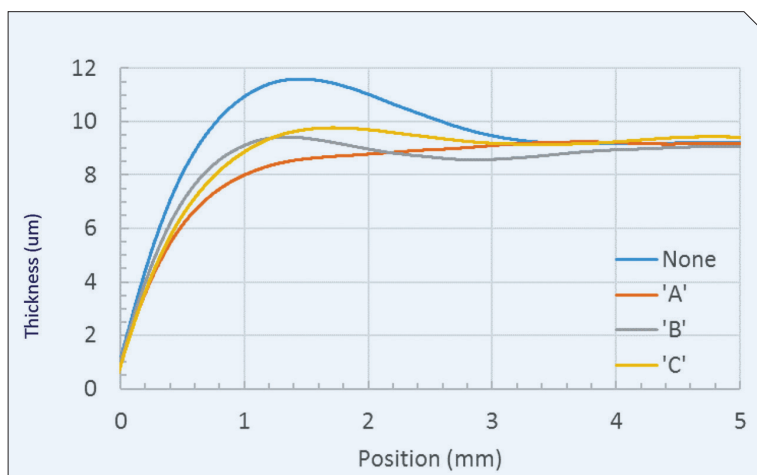
play products on the production line without needing a physical polymer layer mask. Kateeva developed and supports flexible software in which users can easily import CAD files defining arbitrary display shapes including notches and empty areas inside the display areas to be used in recipe generation printing. These shapes are required to support display for features such as camera-in-display or speakers.

In the latest embodiments, it is also possible to print multiple user-controlled thicknesses within one single display subpanel to compensate for topology changes or thickness differences needed to address mechanical stresses in the organic layer. This allows the display to be bent, folded, or rolled. **Fig. 5** shows an example of a (4/8 μm) multithickness planarization layer obtained in a single subpanel print.

Using specially designed software, the polymer edge profile can be tuned by optimizing ink formulation and printing algorithms. **Fig. 6** shows examples of three different edge profiles obtained via different printing algorithms. The edges of 9-μm

Fig. 6.

Fine-tuning the edge profile allowed by edge-compensation software: the blue line is the natural profile, and the A, B, and C profiles are obtained by changing the printing algorithms. The user may select one or the other, depending on the panel design.



thick films acquired by a mechanical profilometer are compared with the natural profile (blue line) defined by the ink rheology and substrate surface energy. Additionally, we can use the physical dam structure around the OLED active area's perimeters to confine the polymer layer.

Choosing Inks

Several factors are relevant in making proper choices on the chemistry to be used for planarization polymer layers. Acrylic and UV curable epoxy monomers and oligomers have been used in formulations that have to meet surface tension and viscosity specifications of printer heads and ink delivery system. Molecular weight, polarity of components and rheological properties of the liquid must match with the surface energy of the surface to be coated for having wettability.

The polymer's chemical interaction with moisture should be considered in balancing the monomer blends' hydrophobic and hydrophilic characteristics to optimize the barrier stacks' morphological and optical stability and their permeation performance. The chemistry also should be compatible with OLEDs' extreme sensitivity to moisture, oxygen, and other small-molecule contaminants. Any volatile or mobile species trapped in the barrier structure could potentially diffuse to the reactive materials in the OLED structure through defects in the inorganic layer(s) driven by a concentration gradient. Any polymer forming via condensation chemistry or containing small-molecule impurities is potentially harmful. Critical contamination may come from moisture contained in the polymer itself and/or residual of a solvent that may be used in the deposition.

Exposure to temperature and UV or particle radiation during inorganic deposition can lead to the organic layers' damage and instability. Morphological changes of poly glycidyl methacrylate (PGMA) films induced by temperature in i-CVD deposition of a SiN_x/organic multilayer barrier has been reported.¹⁸ The same group demonstrated that damage could be avoided by using 1,3,5-trivinyl-1,3,5-trimethyl cyclotrisiloxane (V3D3). Others have discussed the effect of plasma damage to acrylate blends deposited by vacuum polymer deposition in a multilayer sputtered oxide and organic barrier.^{19,20} The damage induced by particle bombardment can be avoided by selected chemistry or by modified processes for inorganic depositions.

The leading supplier of inks for inkjet TFE is Samsung SDI that supplies more than 90 percent of the market. Following the technology's success, major suppliers to the display material market mainly in Japan, but also elsewhere, are approaching the market and are in different stages of engagement with display makers. Smaller startups in the US and Asia also propose new materials. The range proposed covers inks with properties resembling the current ones, as well as functional materials with different mechanical, optical, and electrical characteristics.

Considering Other IJP Polymeric Layers

It is worth mentioning that TFE is not exclusive to flexible displays, but it has been proposed since its inception and is rumored to become commercial for rigid OLED displays to reduce thickness, weight, and eventually fabrication costs. The use of planar polymer layers is also in development for other uses such as functional layers directly deposited on the TFE itself or, on the display during its fabrication. Examples of such layers are the insulator layer in integrated touchscreens for foldable displays²¹ as well as light-extraction features either fully planar or with planarized structured design (such as microlens arrays [MLA]) or as planarization layers in color-on-encapsulation (COE) stacks as alternative to a polarizer to reduce reflected light.³

Planarization layers also are used in OLED TV manufacturing in rigid formats with a double function of planarization and light extraction. Such layers have been deposited by screen printing, but IJP has been proposed and evaluated by major display makers to reduce the displays' material usage and thickness.

TFE has allowed a full exploitation of the intrinsic advantages of OLED technology in flexible displays, thereby enabling their commercialization in mobile applications. Using a polymeric planarization layer between inorganic layers is the key factor in realizing robust TFE technology that meets mass manufacturing and commercial requirements. Looking ahead, liquid deposition by IJP is the technology of choice for polymeric layers' deposition—because of good planarization, easy patterning ability, high throughput, and optimum materials' use. 

References

- Li R. Future of mobile AMOLED market. DSCC/SID Business Conference. 2020 Aug. 3. Available from: <https://www.displaysupplychain.com/speakers/rita-li>.
- Hsiei D. Top 10 display topics in 2020–2021 after COVID-19 & beyond. DSCC/SID Business Conference. 2020 Aug. 3. Available from: <https://www.displaysupplychain.com/speakers/david-hsieh>.
- Young R. Foldable market outlook. DSCC/SID Business Conference. 2020 Aug. 3. Available from: <https://www.displaysupplychain.com/speakers/ross-young-fmo>.
- Moro L et al. Process and design of a multilayer thin film encapsulation of passive matrix OLED displays. Proceedings SPIE 5214—The International Society for Optical Engineering (Organic Light-Emitting Materials and Devices VII). 2003, San Diego, CA. Bellingham, WA: SPIE; c2004. Available from: <https://doi.org/10.1117/12.506549>.
- Moro L et al. A mass manufacturing process for Barix encapsulation of OLED displays: a reduced number of dyads, higher throughput and 1.5 mm edge seal. Proceedings of the International Meeting on Information Display and International Display Manufacturing Conference (IMID/IDMC), Digest of Technical Papers. 2006 Aug. 22–25, Daegu, South Korea. Korean Information Display Society (KIDS); c2006. 754–758.
- Moro L et al. Barrier film development for flexible OLED. Adachi C., Hattori R., Kaji H., Tsujimura T., eds. Handbook of Organic Light-Emitting Diodes. Tokyo: Springer; 2018 [p. 24].
- Graff GL et al. Mechanisms of vapor permeation through multilayer barrier films: lag time versus equilibrium permeation. Journal of Applied Physics. 2004;96(4):1840–1849.
- Aresta G et al. Evidence of the filling of nanoporosity in SiO₂-like layers by an initiated-CVD monomer. Microporous Mesoporous Mater. 2012;151:434–439.
- Ghosh A et al. Thin-film encapsulation of organic light emitting devices. Applied Physics Letters. 2005;86(22):223503.
- Lau KKS, Gleason KK. Initiated chemical vapor deposition (iCVD)

of poly(alkyl acrylates): an experimental study. Macromolecules. 2006;39(10):3688–3694.

- Zambov L et al. Advanced roll-to-roll plasma-enhanced CVD silicon carbide barrier technology for protection from detrimental gases. Chemical Vapor Deposition. 2011;17(7–9):253–260.
- Nominanda H et al. Large area thin film encapsulation from bendable to rollable and foldable. SID Symposium Digest of Technical Papers (SID Digest). 2018;19(1):1103–1105.
- Hong S et al. Development of Commercial Flexible AMOLEDs. SID Digest. 2014;45(1):334–337.
- Graff GL et al. Environmental barrier material for organic light emitting device and method of making. US Patent 6,522,067 B1. 2003.
- Hemerik M et al. Lifetime of thin-film encapsulation and its impact on OLED device performance. SID Digest. 2006;37(1):1571.
- Lin S et al. Ultra-barrier coatings enabled by inkjet printing. 855 American Chemical Society, Division of Polymeric Materials: Science and Engineering, Spring 2010, San Francisco, California, USA, 2010 Mar. 21–25. PMSE Preprints. Volume 102. <http://toc.proceedings.com/07751webtoc.pdf>
- Samsung/SNU Precision. Samsung sector update. 2011 Dec 8. Samsung. Available from: 75303064-Amoled-Overweight.pdf (dday.it)
- Spee DA et al. All hot wire CVD organic/inorganic hybrid barrier layers for thin film encapsulation. MRS Proc 1447. 2012, mrs12-1447-v08-03. Cambridge University Press. Available from: <https://doi.org/10.1557/opl.2012.1065>.
- Moro L, Krajewski TL. Encapsulated devices and method of making. US Patent 7,767,498. 2010.
- Moro L, Chu X. Multilayer barrier stacks and methods of making multilayer barrier stacks, US Patent 7,648,925. 2010 Jan. 19.
- Kang J. AMOLED & foldable display technology trend [presentation]. IHS Markit 38th Display Japan Forum. Tokyo, Japan. 2020 Jan. 30–31. Available from: <https://omdia.tech.informatica.com/OM006350/AMOLED--Foldable-Display-Technology-Trend>.



Lorenza Moro is senior technical advisor at Kateeva Inc. and a technical consultant in the display industry. Moro's previous positions in R&D direction and management were with Kateeva, Samsung, SRI International, Vitex Systems, Inc., and Honeywell. Moro

holds a doctorate in physics from the University of Padova (Italy). Moro is author of about 100 publications in referred journals and scientific meetings proceedings, contributor to numerous books in the field of display technology, and holds numerous US and international patents in the field of electronics materials, processes, and hardware. Reach her at lorenzatest@gmail.com.

Christopher Hauf, an inkjet and color/imaging science expert, leads the team that develops the core algorithms and inkjet writing systems that uniquely distinguish Kateeva's printing systems. He is a key architect of Kateeva's proprietary SmartMixing software technologies. He joined Kateeva in 2012. Before Kateeva, Hauf spent two decades with the Eastman Kodak Company. Hauf received a B.S. degree in imaging science and an M.S. degree in color science, both from the Rochester Institute of Technology in Rochester, New York. He holds both US and international patents related to various inkjet writing systems concepts and algorithms. Reach him at chauf@kateeva.com.

