Roll to Roll Processing for Flexible Electronics

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Abstract: This review discusses the current state-of-the-art technology behind the developing field of roll-to-roll-processing for flexible electronics. The aim is to present a realistic view of the technical obstacles still to be overcome in this field and entertain different ideas on how to achieve them. The need for new substrate materials, innovative patterning methods and impermeable barrier layers are presented in as much detail as possible. Industry perspectives on the cost and product development requirements are also discussed.

Introduction

It doesn't take a large stretch of the imagination to realize that electronics are everywhere. From calculators to laptop computers, we have become accustomed to electronic devices that get more portable every year due to advances in circuit, chip and battery design. The workhorse behind the technology is a technique called photolithography, which, since its invention in 1959, has shrunk the size of printed circuit features 400 fold.[28] However, lithography is expensive because, at high resolution, it can only pattern small areas at a time. The electronics industry is now looking to fabricate structures on a larger scale, and would prefer to use flexible substrates that would have a lower profile, lighter weight and be more rugged than Silicon substrates. Their use would also offer manufacturers the potential for continuous, high throughput printing on large rolls (often called a "web") rather than batch processing small 12′′ wafers. Together, with the rise of organic electronics, "Roll to Roll" (R2R) processing could revolutionize large area electronics manufacturing and could be used to mass produce photovoltaic roofing panels, large area solid state lighting devices, x-ray imagers and flexible flat panel displays.

The flexible display market is expected to grow from $5 million in 2006 to $339 million in 2013, or 83.5 percent per year, according to market research firm iSuppli Corp.[7] Despite the attractiveness of flexible electronics, this new technology must overcome significant technical and process challenges in order to gain practical, high volume applications. Chief among these challenges are those that relate to the cost and performance of flexible circuits, panel size, process throughput, substrate distortion, barrier layer technology and yield. This paper aims to elucidate the requirements necessary for the adoption of this new technology into mainstream manufacturing and discuss the enabling technologies behind the R2R process flow: flexible substrate materials, novel patterning techniques, and robust barrier layer packaging.

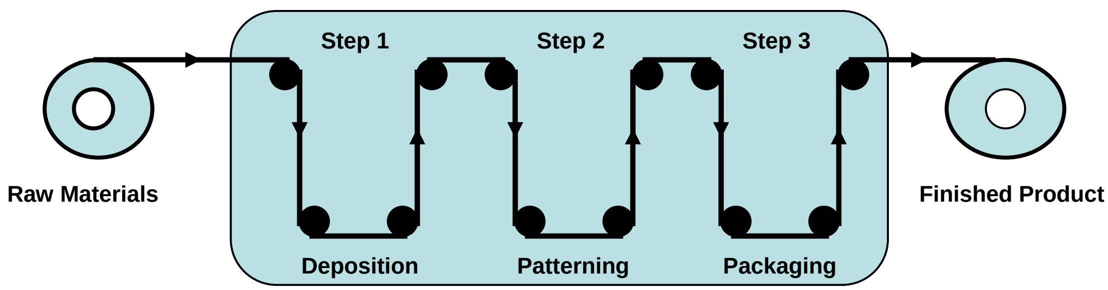

Figure 1. Roll-to-roll manufacturing process flow. Adapted from ref. 24.

A Typical R2R Process Flow

Before moving into industrial-level requirements for the adoption of R2R processing, it will be helpful to describe and define the elements of a typical process flow for a typical flexible electronics process, namely, the creation of a thin film transistor. As shown in Figure 1, the three essential steps of R2R manufacturing are deposition, patterning and packaging. Following the bottom contact TFT device structure shown in Figure 2, a transparent conducting oxide film is deposited on top of the flexible substrate to act as the gate electrode for the TFT. Indium Tin Oxide (ITO) is the current industry favorite due to its superior environmental stability, low electrical resistivity (1 to 3 x 10^{-4} Ω-cm) and high transparency to visible light (>90% at a film thickness of 100 nm).[17] In the interest of space, deposition methods will not be discussed in this review. In subsequent patterning steps, a thin insulating dielectric film of SiO_2 and the metallic source and drain electrodes are produced before printing the organic semiconductor layer. Rather than conventional photolithography methods, soft lithography methods, laser ablation, and inkjet printing methods seem like promising technologies for large area flexible displays. Finally, the devices must be packaged in a barrier layer to prevent oxygen and moisture contamination of the organic semiconductor layers.

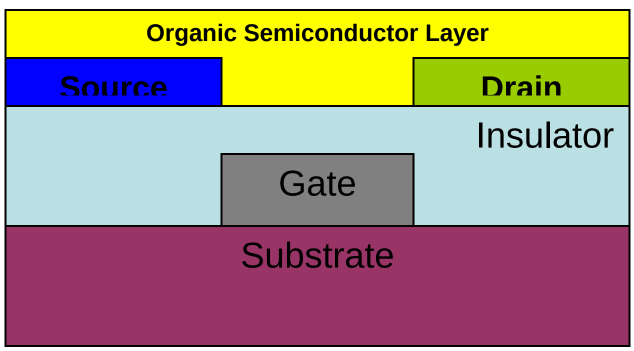

Figure 2. Schematic device structure of a bottom contact organic thin film transistor.

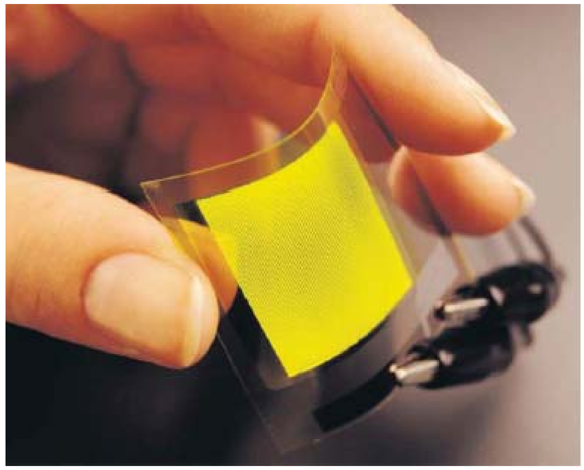

Figure 3. Photograph of an OLED display. Courtesy of DuPont.[10]

Substrate Materials

The lack of a clear choice for substrate material is inhibiting the development of flexible displays. The ideal substrate would be transparent (for OLED emissive displays), flexible and rollable, low-cost, resistant to chemical attack, dimensionally stable under thermal cycling and would have low permeability to water and oxygen and thus able to act as an intrinsic barrier layer. If this barrier layer were inherent in the substrate, simply laminating two film sheets together would be sufficient to package the device at low cost. No material has emerged that fills all of these needs (see Table 1), but the three that have come close are glass, plastic and stainless steel.

Property	**Stainless-Steel**	**Plastics (PEN, PI)**	**Glass**
Thickness (μm)	100	100	100
Weight (g/m^2)	800	120	220
Safe bending radius (cm)	4	4	40
RTR processable?	Yes	Likely	Unlikely
Visually transparent?	No	Some	Yes
Max process temp (°C)	1000	180, 300	600
CTE (ppm/°C)	10	16	5
Elastic Modulus (GPa)	200	5	70
Permeable O_2, H_20	No	Yes	No
Pre-bake required?	No	Yes	Maybe
Planarization necessary?	Yes	Maybe	No
Electrical conductivity	High	None	None

Table 1. Comparison of substrates for flexible electronics, from ref. 16.

Substrate Materials: Glass

Thin sheets of glass can be made flexible with very small thicknesses (less than 100 μm). They are the standard substrate material at the moment, acting as perfect impermeable barrier layers and offering superior optical properties, ultra-smooth surfaces, and low thermal expansion coefficients. However, such thin sheets of glass are highly susceptible to breaking and cracking along the edges if even slightly mishandled. Coating the glass sheets with a thin polymer layer around the edges and surface makes the substrates less prone to breaking during minor handling mistakes in production and also reduces the influence of already existing defects. Even with this hybrid approach, glass substrates cannot currently be rolled up due to low yield related to glass breakage. [11] It may be possible to use an ion-exchange process to strengthen the glass and reduce breakage, however. By simply immersing a glass sheet in a molten bath of silver nitrate, small sodium ions in the glass matrix are replaced with larger silver ions through an interdiffusion process. Similar to thermal tempering, this results in compression on the glass surface and tension in the interior. More work in this area is necessary in order to make glass a viable substrate for R2R processing.

Figure 4. Photograph showing thin, flexible glass. [26]

Plastic Films

Plastic engineered films are very appealing substrate materials for flexible electronics due to their low cost and toughness, of which Dupont's Teonex® brand of polyethylene naphthalate (PEN), is a leading candidate. As seen in Figure 5, PEN shows a remarkably smooth, defect-free surface quality after pretreatment with an adhesion layer, but has a Young's Modulus three times higher than amorphous plastic films due to its semicrystalline, biaxially oriented nature. This stiffness may hinder PEN's placement into an R2R processing format.

Substrates for flexible electronics must be able to withstand temperature cycles required for the deposition of barrier and indium tin oxide coatings. During these heating cycles that can reach temperatures over 250 °C, plastic films such as PEN undergo a molecular relaxation process at a certain temperature (different for each polymer) called the glass transition temperature (T_g), which alters the physical and mechanical properties of the film. At temperatures near the T_g the polymer starts to flow like a liquid and the dimensional stability of the material under tension drops dramatically. Due to its T_g at ~120 °C, its unlikely that PEN will be compatible with current high temperature ITO deposition methods.

Shrinkage is a factor in semicrystalline substrates such as PEN. During the manufacturing of the film, the polymer chains become biaxially oriented along the direction of the machine rolling process. Upon cooling of the polymer film after the ITO deposition step, the films shrink back towards their original dimensions along the machined direction. Shrinkage of more than 2 percent may cause cracking in top layers of the ITO conducting electrode material and destroy the properties of the flexible display. [10] However, pre-annealing the film under high temperatures and minimal web tension can enhance the dimensional stability of the film by counteracting the effects discussed above. This 'heat-stabilization' process relaxes the internal strain induced by the manufacturing process and allows the polymer chains to reorient themselves in a more stable configuration. Afterwards, the dimensional change in the substrate becomes much more predictable. [14] As a result, it may be possible to design alignment software with appropriate models to adapt to the dimensional changes in the plastic substrate.

Natural thermal expansion also needs to be taken into account when dealing with thin film stacks on polymers. Thermal expansion is based on a number called coefficient of thermal expansion (CTE) of the material. Relative to other polymers, PEN has a relatively low CTE of about 13 ppm/ °C, but what is important is that the substrate is coupled with a layer that has a similar CTE. [10] Any mismatch in thermal expansion coefficients during thermal cycling could cause high levels of residual stress and film cracking.

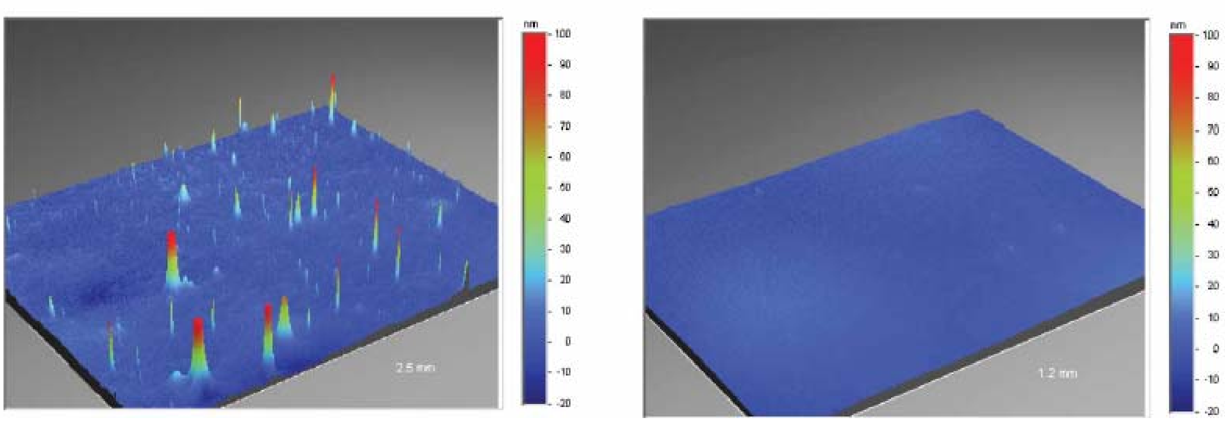

Figure 5. Atomic force microscope height images showing the surface smoothness of a) industrial grade polyethylene napthalate (PEN) and b) DuPont's surface tailored Teonex® Q65 film, from ref. 10.

Stainless Steel

Stainless steel (SS) is a leading substrate candidate for applications where transparency is not required. SS foils, produced with thicknesses of 125 μm, provide a durable, flexible substrate that tolerates high temperature processes with much better dimensional stability than plastic. The foils provide a perfect diffusion barrier to oxygen and water vapor and have proved to be successful substrates for both amorphous and crystalline Silicon based TFT's used to make top-emitting active matrix OLED devices.[12] Finally, at 10 ppm/ °C, SS films also have a lower CTE than polymer films.

The steel foils, however, have a rough surface due to rolling mill marks and are highly conductive, so an insulating spin-on-glass (SOG) planarization layer must be coated on top of the stainless steel. This ensures flat, non-conductive surfaces that will translate into accurate device registration on subsequent layers. These layers add

~$20/m^2 to the cost of using stainless steel as a substrate, but they are still estimated to cost less than plastic substrates. Plastic substrates for AMOLED's require multilayer barrier coatings estimated to cost greater than $50/m^2, while stainless steel substrates need only one chemically vapor deposited coat of insulating amorphous Si.[13] Stainless steel substrates look like very promising candidates for R2R processable devices.

Figure 6. Photograph showing United- Solar Ovonics solar cells on flexible stainless steel. The Ovonics solar cell line may be considered the first mass produced roll-to-roll process application, from ref. 13.

Novel Patterning Techniques: Contact "Soft" Lithography

Researchers in the field have come up with innovative techniques to try and bring the cost of R2R patterning processes down to a reasonable level. Abbie Gregg Inc., a consulting firm based out of Tempe, AZ report calculated that materials costs comprise over 50% of the total active matrix OLED factory cost in a roll-to-roll process factory.[24] This number is calculated based on the assumption that photolithography is used for device patterning. The multiple photoresist applications and etching steps involved in photolithography dominates this material cost.[2] Newly patented "Self-Aligned Imprint Lithography (SAIL)," would eliminate multiple photoresist applications. In SAIL, multiple mask levels are imprinted as a single three dimensional structure. The photopolymer layer is heated above its glass transition temperature to allow it to flow into the crevices of the stamp. The stamp/ polymer sandwich is cured with UV light as the polymer cools and hardens, allowing the stamp to pull off cleanly. The process is completed with standard wet and dry etch processes, leaving an accurately reproduced 3-D, high resolution pattern on the substrate. The technology is called self-aligning because the mask would deform with the substrate during the embossing heat treatment step.[6]

At issue with "soft lithographies" such as SAIL is that the contact between the mask and the substrate increases the likelihood of defects due to particle contamination. Also, mask damage and the lifetime of the flouro-elastomer stamping materials raises doubts about the robustness of the process. However, one research group has shown that the defect numbers actually decrease during use, indicating a "self-cleaning" process.[4]

Tan et. al. has developed roller-type nanoimprint lithography, which has a lighter, more uniform imprint force along the contact area. It also features a simpler design construction that is able to repeat a mask continuously on a large substrate. This technique, similar to gravure printing, seems more naturally suited to the R2R process flow.[5]

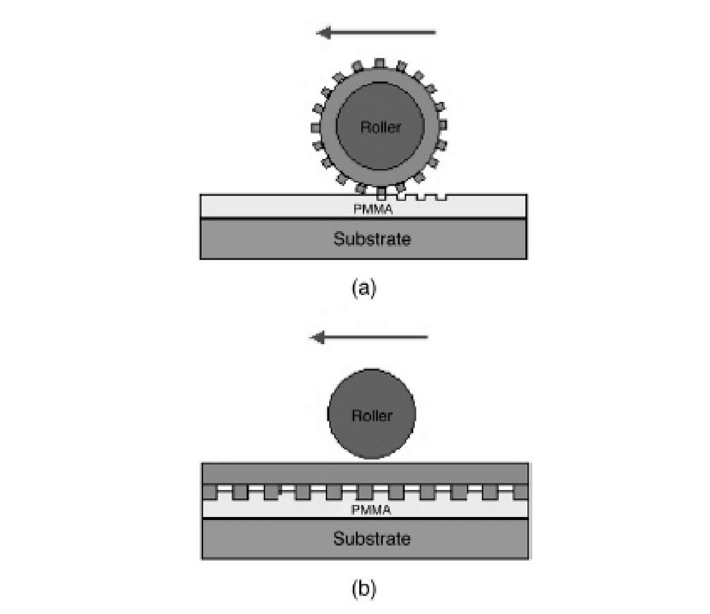

Figure 6. Shows two methods of roller nanoimprinting: a) mounting a stamp around a cylindrical roller and rolling onto the substrate, b) putting the mold directly on the substrate and rotating the roller on top of the mold. Image reproduced from ref. 5.

Novel Patterning Techniques: Laser Ablation

A technique that would eliminate both the photoresist coating and wet etching steps is called laser photoablation. This technique is used to write directly write into a polymer layer using a high powered laser. As shown in Figure 7, photoablation works by breaking molecular bonds in a polymer layer, fracturing the polymer into shorter units that are "kinetically ejected" upon removal.[3] The amount of material ejected can be tuned by adjusting the wavelength, energy density and pulsewidth of the XeF excimer laser used for ablation and is capable of reproducing ablation depth to within 0.1 μm across large areas of the substrate. Examples of ablatable polymers are polyimides (e.g “Kapton”) and PET (e.g Mylar), which are also commonly used substrates in flexible electronics.

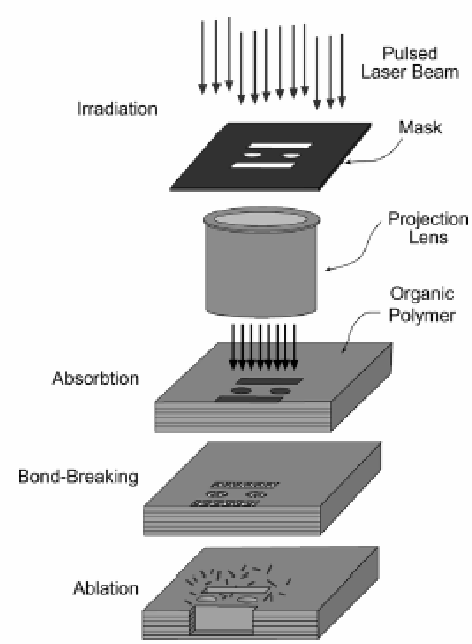

Figure 7. Schematic of the laser ablation process. Image reproduced from ref. 27.

A R2R manufacturing line can only be classified as high throughput if all of its component processes are performed at high throughput rates. In other words, a manufacturing production line is only as fast as its slowest process; bottlenecks must be avoided at all costs. Traditional material removal processes to create lines and vias in electronic packaging include techniques such as reactive ion etching (RIE), thermal, and mechanical drilling. According to Kanti Jain of Anvik Co., photoablation can be significantly faster than RIE and produce cleaner lines than thermal and mechanical drilling. His group has developed a high speed, large area photoablation process that uses a seamless serial scanning technique to produce patterns on a copper-on-Kapton substrate.[3] Supply and take-up rollers maintain the tension of the web without creasing or kinking as it moves through the exposure area. The web is held down during exposure to avoid image distortion by a vacuum diffuser, which is a smooth, porous material. The vacuum diffuser ensures that no web deformation takes place during the exposure. After the exposure is complete, the vacuum chuck releases and the web is rolled to the next segment. The system is capable of a resolution of 10 microns and an exposure throughput of about 120 panels per hour, or 6 square feet per minute.

Novel Patterning Techniques: Inkjet Printing

While laser ablation may be called a subtractive technique, inkjet printing can be considered an additive technique. Rather than your home, graphics-oriented inkjet printer, an array of piezoelectric printheads will be required for depositing conducting organic solutions at precise locations. Litrex (Pleasanton, CA) now has a Generation 8 system up and running that can achieve resolutions of up to 120 points per inch, droplet sizes ranging from 10 to 80 picoliters, and speeds of up to 200 millimeters per second. Each droplet is monitored by high speed camera technology, which ensures that each nozzle is working as it should.[31] It may also be possible to print "slurries" of Silver or

Zinc Oxide nanoparticles to form conducting patterns, which, when “sintered” have mobilities greater than 10 cm^2 / V-sec, much better than any organic material![30] Inket printing, however, currently suffers from a considerable error in drop placement accuracy of +/- 10 μm. Pre-patterning hydrophilic and hydrophobic regions onto the substrate before inkjet deposition may be a more precise way of ensuring that the droplets spread exactly where they are needed. Engineers are also working towards a solution for the “coffee stain effect” in which organic solute is transported to the rim of the droplet upon evaporation.[32] These problems emphasize the fact that many different types of scientists, such as fluid dynamics experts, chemical, electronic and materials science engineers, will be required to solve technical problems in this field.

Before insertion into a patterning process flow, further studies of each of these techniques are required to weigh their maximum achievable pattern registration versus cost. In the end, many different technologies may make it to the factory floor, depending on the specific needs of the application. Photolithographic techniques will arrive much later, when very high resolutions (< 100 nm) are required and resist, development, and etching steps become compatible with plastic substrates.

Barrier Layer Technology

OLED devices will not perform without first encapsulating them in a moisture and oxygen free environment. The reason for this is that the low work function metals used in OLED's oxidize very easily, creating non-emissive black spots on the organic emissive layer and ruining the display. The benchmark required for long device lifetimes are water transmission rates of less than 10^{-6} g m^{-2} per day and oxygen transmission rates of less than 10^{-5} mL m^{-2} per day. In order to test state-of-the-art devices, the current method used is to hermetically encapsulate the device in a thick "can" of epoxy sealant and glass that is three times thicker than the device itself.[15] Even with this technique, oxygen may diffuse through the epoxy and device lifetimes may only last several hours. The best thin film packaging methods have demonstrated water vapor permeation rates of ~0.5-1.0 g/m^2 per day, which is 3-4 orders of magnitude above the benchmark value.[21] The market is waiting for a thin film barrier layer coating that will work to permanently keep oxygen and moisture away from the active elements of the OLED device.

The leading candidate to realize these strict requirements is called Barix® and is composed simply of many alternating layers of polymer and ceramic, adding up to about 3 microns of thickness to the device (Figure 8). The idea behind this technology is that even near perfect moisture barrier films such as vacuum deposited, sputter coated SiO_2 will crystallize in micro columns, creating variations in density and allowing oxygen molecules to diffuse easily between the columns. Pinholes are also present in such films, allowing oxygen and moisture seepage. Barix® solves this problem by using continuous, roll to roll vacuum processing techniques to stack multiple ceramic films on top of polymer isolation layers that provides a torturous diffusion path for the oxygen molecules that is almost too slow to measure with current technology.[21]

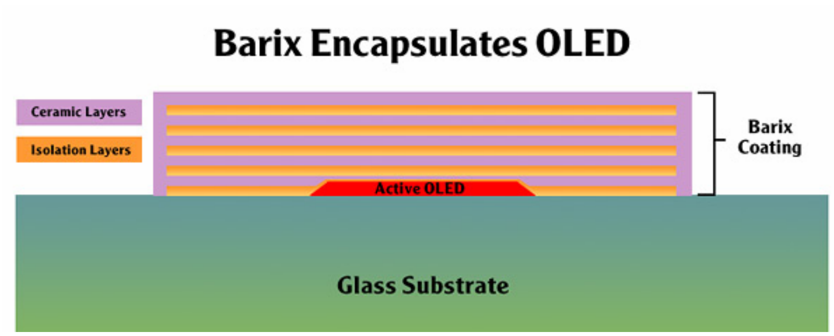

Figure 8. Schematic of Barix barrier layer technology deposited on a glass substrate. From http://www.vitexsys.com/encapsulation.html.

Industry Perspectives: Cost

In order to succeed as a viable manufacturing alternative, roll-to-roll processing needs to show a dramatic reduction in cost compared to the current technology. The infrastructure for manufacturing large area flexible displays does not yet exist, so factories wishing to incorporate R2R processing technologies would have to deal with very high start-up costs due to custom-built tools. "In order to compete with existing technologies, flexible flat panel displays must beat the current technology in cost by about a factor of four," according to Professor Michael Thompson, former co-founder of a flexible electronics startup Flexics Inc. A company willing to enter the flexible electronics market will need this type of economy to guarantee that their initial start up investment will pay off in the long term. For example, current 20'' flat panel monitors cost around $550, putting the price at around $1.25 per square inch. Therefore, the price of a flexible flat panel display would have to sell at around $0.30 per square inch in order to be marketable in a roll-to-roll process format. However, the recent (2003) cost model presented by consulting firm Abbie Gregg Inc. placed the current cost of setting up an R2R factory capable of designing active-matrix OLED displays at around $1.23 per square inch,[24] which does not show enough savings for a green light in manufacturing, not to mention the other current technical challenges in creating OLED devices that have not yet been solved.

Figure 9. Photograph of "smart card" application. The display is created using E-ink® electrophoretic technology (www.e-ink.com).

Industry Perspectives: Product Development

The market is currently waiting for a "killer application" in order to justify the entry of widespread R2R into electronics. This application must not only be more economical, but should be able to tolerate a high defect density, being subject to inevitably low process yields. The application should also have broad appeal in the business or consumer market, rather than in specific medical, military or government niches. [33]

“Smart cards,” as shown in Figure 9, are flexible credit cards with password activated displays that could show declining balances and other card-related information. These could be a potential application, but how much flexibility is really required on a credit card? A similar effect could probably be accomplished with the implantation of a small silicon chip and plastic packaged LCD screen into a normal credit card. Even better would be an entirely new application that utilizes the entire area of the device, and satisfies a consumer need that no other device can touch.[9]

Industry leaders are still shaking their heads over what this “killer app” should be. The result is a real “chicken and egg” problem, according to Mark Strnad, Abbie Gregg’s vice president of operations and engineering. Display makers rely on production tool makers to custom build equipment. At the same time, tool makers continue waiting for the display makers to define the form and function of the production tools required.[1] Industry is feeling the effects of this lack of progress, and ultimately led to the demise of the OLED research group at Hewlett Packard.[30]

Excitement over flexible technology has spurred several start-up companies to get involved in the flexible display market and large companies such as General Electric have large internal research contracts to study and develop the technology.[22, 23] Government sponsorships, such as DARPA, are spending heavily on flexible display technology for the “Future Force Warrior,” who will need flexible, portable and rugged maps of battle terrain.[8] DARPA also spends heavily on “smart camouflage," flexible displays that conform to an aircraft wing and change colors to match their environment. However, none of these parties have emerged as the "champion" of flexible displays, according to Kimberly Allen of iSuppli Corp.[33] This champion would act as a catalyst to generate excitement in potential customers and partners, as well as inspire fear of being left behind among its competitors. Government and academic groups are sufficient leaders of the industry at the current research and development stage, but significant corporate level funding is needed to jumpstart an industry and build factories. Until such a champion appears, it is hard to imagine the flexible electronics market getting off the ground.

Figure 10. A prototype model of a potential flexible flat panel display application. The device could be rolled up into a pen when not in use. Images courtesy Universal Display Corp, from ref. 25.

Conclusion

The purpose of this review was to discuss the current status of the roll-to-roll processing for flexible electronics market. The review only skimmed the surface of the many promising technologies that could contribute to this field in the future, and the reader is encouraged to look up the references below for more information.

Although the field of flexible electronics has made significant advances in technical and process capabilities in recent years, much more work needs to be done before the field is ready to be scaled up for R2R process technology. It is not a given that roll-to-roll processing will ever come to full fruition. If the technology matures as hoped within the next decade, new substrate materials and cheap, innovative patterning techniques must be combined with creative product development teams to come up with compelling flexible electronics applications. The combination of novel consumer products and the ability to manufacture them using an ultra low cost production capability such as roll to roll processing will entice an adventurous company to take the risk and invest in a full-scale flexible electronics factory. Other companies will follow suit with their own innovations, and a revolution in electronics manufacturing will begin…or will it?

Acknowledgements

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