Polyester Film Substrates for the Flexible Electronics Industry - An Overview and Where Next

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1. Introduction

Since the early 2000’s there has been an explosion in interest in printed electronics both in terms of developing the technology and also in exploring the potential to develop new business opportunities. The opportunity to exploit flexible substrates in R2R production has been one that has excited the interest of the plastic films and associated processing and coating industries. During the past decade a considerable network of activities based on companies developing new technologies, and established companies from within the potential supply chain have emerged. It is now possible to compare and contrast the vision of what the early thoughts on material requirements were and how the technology might be exploited, with what has emerged.

(i) Physical Form/Manufacturing Route: Initially the potential to prepare displays based on a flexible substrate opened up the possibility of thin, robust, rollable, colour displays based on OLED technology, but the technical difficulties in developing a high barrier on a flexible substrate has limited the development of flexible colour displays. The prototype rollable and flexible displays sampled to the market based on electrophoretic black and white displays were superseded by the introduction of the smart phones and tablet based devices; the public now expects the sophistication of full colour displays, and public acceptance of the tablet form has negated any desire for a rollable display concept. Despite this and the fact that present day displays remain largely flat, ruggedness and light weight remain desirable attributes that displays on flexible substrates can offer. In terms of manufacture, the vision of a roll of film at one end and devices emerging at the other end was postulated. Although R2R is used extensively for specific steps in substrate production including film manufacture, conductive coatings and barrier coatings, device manufacture is still largely batch based, partly because this fits best with the semiconductor industries existing manufacturing tooling equipment.

(ii) Film in Use: Films now find use in a much broader range of application areas than the OLED displays that were originally envisaged eg printed circuitry, printed memory, electrophoretic displays, sensors, OLED lighting and flexible PV devices. In addition flexible film is used extensively in rigid electronic devices eg in touchscreen and in light management films in LCD displays. This has influenced the focus on flexible substrate development over the past decade and this talk will initially give an overview of the evolution of substrates of different types suitable for flexible electronics, exploring the issues associated with substrate development and selection. The talk will then lead on to discuss the latest film developments in DTF in support of the flexible electronic and PV industry.

2. The Flexible Substrate Property Challenge
2.1 Substrate

Over a decade ago dimensional reproducibility at processing temperatures, surface smoothness, high barrier and inertness to the chemicals used in processing were highlighted as the key property requirements. Cost and commercial availability have emerged as further film requirements as manufacture has moved from demonstrators to prototypes to commercial manufacture with the realisation that only film manufactured at a commercial scale is likely to meet the consistency and quality required for commercial device manufacture. At the current time figure 1 shows the main plastic film types under consideration by the flexible electronics industries.

![Figure 1 Films regularly referenced as substrates for flexible electronics](image)

The films can be categorised into semicrystalline and amorphous. The combination of dimensional reproducibility, surface quality, chemical inertness cost and commercial availability that have given biaxially oriented, heat stabilised polyester films a leading position in this emerging market have been discussed elsewhere in more detail. The films that are circled have emerged as substrates of interest over the past decade. Oriented polypropylene film (O-PP) is finding use in “simple” printed electronic applications where processing and performance at higher temperatures is not required. Polyether ether ketone (PEEK) is the premier high performance semicrystalline engineering polymer and offers an excellent high temperature, chemical inertness property set. The amorphous high Tg resin developed by Akron Polymer Systems offers high temperature processing. However new polymer and or film developments have the issue of the prohibitive costs of scaling up to a commercial quality film line for what remains at present a low volume market. Other materials such as stainless steel and flexible glass have also been worked on. Stainless steel finds use in PV devices, but is not being actively used in flexible electronics. Flexible glass offers transparency and excellent barrier properties, but faces the challenges of scale up to a commercial scale, and technical issues associated with edge cracking.
Experience over the past decade has shown that material “off the shelf” is unlikely to have the complete desired property set required, but good progress can be achieved when the device manufacturer and the substrate supplier work together to match process capability to an optimised substrate property set. Taking the property set highlighted above, and using the biaxially oriented polyester films polyethylene terephthalate (PET) and polyethylene naphthalate (PEN) as examples, it is possible to review the progress that has been achieved.

(i) Flexible Transparent High Barrier

Although considerable progress has been made in developing high barrier films and commercial products are now available, achieving a cost effective barrier of $>10^{-5}$ g/m2/day for moisture and $>10^{-5}$ mL/m2/day remains a key challenge and a rate limiting step in developing commercial quality flexible OLED displays. A pristine film surface is one of the key enablers to achieving this and progress towards this will be discussed later.

(ii) Dimensionally Reproducibility

Melinex® ST504/506 and Teonex® Q65HA family of films exhibit dimensional stability up to approximately 150°C and 180-200°C respectively\textsuperscript{1,2,3,4}. The dimensional reproducibility of the films can be enhanced further by the use of a rigid carrier, the use of process technology that allows for dimensional change and control of the processing atmosphere (minimising the effects of swelling due to moisture pickup). Once a certain level of shrinkage is achieved consistent dimensional reproducibility is more important than further reductions in shrinkage. Companies such as Plastic Logic\textsuperscript{TM}, Polymer Vision\textsuperscript{TM} and ASU Flexible Display Center have demonstrated the successful manufacture of TFT backplanes on polyester film evidencing that the required dimensional reproducibility can be achieved

(iii) Surface Quality

Defects on the surface of the film are clearly undesirable and can lead to pinhole defects in barrier coatings, or line outs and spot defects in displays. It is probably impossible to achieve glass like perfection in terms of surface smoothness with a plastic film. It can be argued however, that perfection is not actually required. What is important instead is to understand what defects are critical to device manufacture and to target and eliminate these defects where possible in a cost effective manner. This is an area that DTF has researched extensively and continues to be active in. Experience over the past decade has shown that external debris (dust, externally introduced debris, scratches etc) rather than internal contamination is the major issue and one approach adopted to overcome this has been to cover any defects or marks on the surface with a planarising coating; it has been demonstrated that the introduction of such coatings significantly reduces the defect count on the surface\textsuperscript{3,4}.

White light interferometry has been used extensively to character the surface of the films, but the use of Roughness Average (Ra) and Root Mean Square Roughness (Rq) can be misleading as they tend to be measured over a small sampling area and do not capture the occasional peaks that occurs on the sample size of a display. DTF have worked with a leading surface metrology tool manufacturer to develop a Large Area Metrology ‘LAM’ tool-the first of its type. This device has the capability to defect map the surface of film fast and precisely, achieving the optimum lateral resolution obtainable from white light interferometry. The equipment was designed in addition to help distinguish between both intrinsic (polymeric based) and extrinsic (external i.e. air borne debris) defects.
Typical output:

(i) Sampling area = 35x35 cm measured in around 2 hours.
(ii) Map of intrinsic and extrinsic defects, precise locations registered for compositional analysis
(iii) Full X'Y'Z measurements of the surface defects, enabling significance testing.

Such output can then be used to identify defects critical to device manufacture, and help determine the impact on the effectiveness of removal strategies. An example of this is where the LAM tool was used in support of a UK Government funded programme ("HighQSurf"), the aim of which was to achieve a defect reduced surface by

(i) characterising the effectiveness of substrate cleaning techniques,
(ii) identifying the defects critical to device manufacture and tracking where the defects were introduced in the manufacture of the film and subsequent handling and transport of film into a device manufacturers facilities
(iii) identifying and applying strategies to reduce defects and improve device manufacturing yields.

DTF worked with one of the project partners, Plastic Logic\textsuperscript{TM}(PL), to identify defects with the potential to cause shorts and line outs etc. and attempted to work back through film handling to identify where the defects were introduced.

The image in Figure 2 was taken from a PL device ‘mask’ – comprising of an array of circuit lines 40um apart, 10um wide and 50nm high. The mask plate was then electrically tested with an X Y grid array to locate the short regions and the short regions were studied using the LAM. Figure 2 shows an example of a particle of extraneous debris giving rise to a short. The defect regions were first of all quantified in terms of their height significance; further analysis could be then carried out to determine the composition of the defect. This particular study revealed that virtually all shorts were due to externally introduced detritus.
Within the “HighQSurf” programme, a major audit of the film entering device manufacturers clean rooms was carried out. The summary conclusions from this audit was that the debris on the surface was derived from human, packaging and process interactions i.e. not the initial film manufacture, but from the subsequent slitting, packaging, handling, transport etc. steps. The conclusion drawn was that there is considerable scope for improving the surface film quality by addressing “hygiene” factors. Attention to detail is one aspect of this, but given that film handling and transport is unavoidable, improved methods of surface cleaning become important. Trials involving Teknek™, another partner within the programme led to the development by Teknek™ of their Nanocleen roller technology with the composition and configuration of rollers designed to remove sub-micron as well as larger particles. Improvements in static control and the provision of a protect film on the surface of the planarised films have led to further improvements in film surface quality and the adoption and exploitation of techniques such as described above have taken Plastic Logic™ to the stage where they now achieve the same yields on polyester film as they achieve on glass.

3. New Film Developments

In addition to above new films are being developed to make further improvements targeting surface cleanliness, but also to address additional film requirements that have arisen in the more recent past

3.1 “On Demand” Clean Film

The polyester film surface on manufacture is almost glass smooth; as discussed above the surface becomes compromised during the subsequent handling and transport processes. One approach adopted by DTF has been to exploit coextrusion technology to coextrude a sacrificial film layer on to the surface of the film during film manufacture. The polyester film surface becomes an internal layer which is only exposed to the environment when the sacrificial film layer is removed, ideally at the point of device manufacture. The sacrificial layer has the added benefit of protecting the surface of the film from contact damage during film handling and transport.

Figure 3 Schematic view of “On Demand” Clean Film

The impact of the sacrificial coextruded layer on subsequent processing has been studied by Dr. Sandeep Unnikrishnan of the TNO-Holst Centre within a European fund FP7 programme CLEAN4YIELD. A proprietary single layer inorganic barrier material was deposited on the surface of the film and a version of the calcium test has been used to study the decay of the calcium on barrier film on regular film surface and a on a film surface after the sacrificial layer has been removed. Qualitatively it can be seen that relative to the control the calcium
decays (as denoted by the Ca test squares becoming lighter) at a much slower rate on the “OnDemand” clean film compared to a normal film surface indicating the impact of the protect film in preserving the polyester film surface quality.

Figure 4 Calcium test pieces aged at 20°C and 50%RH

A further advantage is that the sacrificial coextruded layer protects the PET surface from defects that may arise by contamination build up at the die lip edge and/or when the film passes over rollers during the manufacturing or slitting processes. Molten polymer flowing over any debris build up at the die lip edge can lead to lines of scratches with high aspect ratio. These are absorbed within the sacrificial coextruded layer leaving the PET film surface undamaged.

3.2 Low Bloom Film

PET film contains ca 1.4 weight% cyclic oligomer and a portion of these oligomers can migrate to the surface if the film is held at elevated temperatures for tens of minutes, as can be seen in figure 5 below. This “blooming” effect gives rise to haze. PEN with 0.3 weight % cyclic content has significantly lower cyclic oligomer content compared with PET and there is significantly less “bloom” associated with PEN film.

Planarising coatings or hard coats act as a barrier to cyclic oligomer migration and offer one strategy to reduce the haze associated with this “blooming” effect. Careful control of the
filming process offers another strategy to a “low bloom” film. These low bloom films typically have a haze less than or equal to 1% after ageing at 150°C for 30 minutes. As a further refinement on the low bloom films, refractive index matching coatings are being developed which reduce the iridescence that can be observed with hardcoats and conductive coatings that arise because of a RI mismatch between coatings.

3.3 Light Outcoupling Films

DTF have developed a range of volume scattering film which when applied as an overlaminate to a rigid OLED, will outcouple light trapped in the “glass” of the structure. This is an alternative approach to the familiar microlens array technology, and offers advantages in practical areas of surface performance, appearance and viewing angle distortion.

Figure 6. Angle Dependent Colour/Emission from Glass OLED using DTF Outcoupling Overlaminate (courtesy of Holst Centre, Eindhoven)

Through its participation in the European funded FP7 programme “FLEX-o-FAB”, DTF is combining its experience in optical outcoupling and clean substrate technology with other participants to develop a cost-effective R2R process route to flexible OLED lighting. DTF and Holst have already demonstrated the principle of emission enhancement for flexible OLED using a flexible polyester substrate with integrated volume scattering features. The efficacy increased from 30 lm/W to 42 lm/W by using Holst/DTF barrier foil with integrated outcoupling enhancement.

3.4 Weatherable Films

Polyesters such as PET and to a lesser extent PEN are inherently susceptible to degradation by hydrolysis. However the hydrolysis behaviour of PET film can be improved significantly by control of variables such as molecular weight, the crystallinity developed during the filmung process and by control of end groups. The relative susceptibility of different films to hydrolysis is often studied using a Damp Heat Test (DHT) in a climatic chamber set at 85degC/85%RH. By using strategies such as those outlined above the lifetime to failure in DHT can be pushed from ca 1000 hours for a standard PET film to 2000-3000hours for a hydrolysis stabilised film.

Similarly a standard PET film will show poor stability to UV in outdoor use and information based on unstabilised PET films is often used misleadingly in comparison with other film types. PET film can however be very effectively protected against the effects of UV
irradiation by the addition of well-established UV stabilisers. Figure 7 shows results from our laboratory which illustrates how the addition of a UV stabiliser has a significant effect on the retention of mechanical properties of PET as measured by elongation to break (ETB) on accelerated ageing. (Ageing at 10000 hours in a Weather-Ometer® equates to approximately, 3 years/1000 hrs for Northern Europe, 2 years/1000 hrs for Southern Europe and 1 year / 1000 hrs for Australia). Ongoing development work is extending the lifetime of PET film further.

Figure 7 Change in elongation to break versus hours in a Weather-Ometer® for PET (triangle) and UV stabilised PET (squares)

4. Conclusion

The understanding of the property set required for films in use in flexible electronics applications has moved considerably over the past decades and significant progress has been made in developing films appropriate for use. DTF continues to innovate to anticipate the likely future requirements of both the flat panel display industry and the embryonic flexible electronic, PV and OLED lighting market.

5. References