

# Optimising Polyester Films For Flexible Electronic Applications

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**Keywords :** Polyester films, polyethylene terephthalate, polyethylene naphthalate, flexible electronics

## Abstract

*DuPont Teijin Films (DTF) has developed a family of highly stable engineered substrates specifically for the flexible electronics market. Teonex®PEN films and Melinex® PET films are biaxially oriented semi-crystalline polyesters. The requirements for these films can be demanding as they are replacing rigid glass substrates. It is important to understand and control factors that can influence device properties to achieve the optimum performance for a given flexible display application. Trends in the market requiring thinner substrates with very low, uniform shrinkage, improved flatness, improved cleanliness and extremely smooth surfaces will be covered. The various substrate requirements for different applications and latest examples of film in use will also be highlighted.*

## 1. Introduction

The present interest in flexible electronics in the area of displays is mainly centered around the development of liquid crystal displays (LCDs), organic light emitting diodes (OLEDs) and electrophoretic displays (EPDs). In these applications, the aim is to substitute a thick and rigid glass by a thinner and flexible material. Ideally, it would need to offer the same properties as glass such as clarity, dimensional stability under heat and moisture, thermal stability, good gas barrier, solvent resistance, low thermal expansion and a smooth surface. DuPont Teijin Films (DTF) offer polymer films made of polyethylene terephthalate (PET, Melinex®) and polyethylene naphthalate (PEN, Teonex®). Teonex® Q65 and Melinex® ST506/504 are biaxially oriented semi-crystalline polyesters. The high dimensional stability of the substrates, which is required for display manufacturing, is achieved through off-line stabilization. In this process, the internal strain in the film is relaxed by exposing it to high temperature whilst keeping the line tension low. Both films have a good balance of properties whereas Teonex®Q65FA is

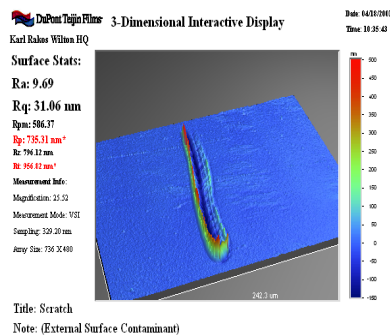
a higher performance substrate that is the preferred choice for higher temperature applications and/or processes [1]-[7].

## 2. Results and discussion

### *Planarised films*

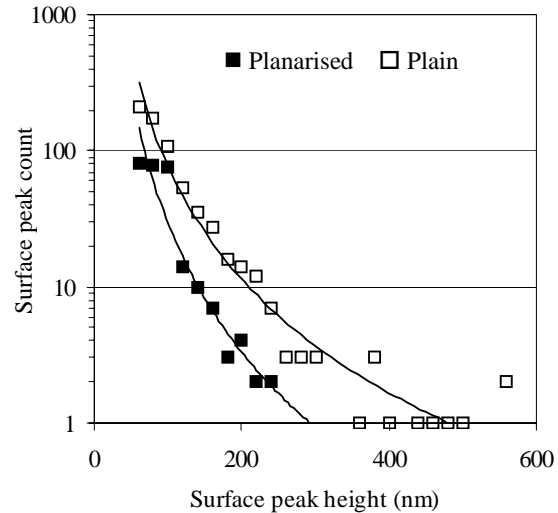
The surface quality of polyester films can be controlled to an extent via recipe control, plant hygiene, web cleaning techniques and film line processing conditions. However, the film still has sporadic surface peaks from internal contamination and is typically not manufactured in an environment which is clean enough to give a level of extrinsic contamination suitable for the more demanding applications in flexible electronics. While a number of cleaning techniques can reduce extrinsic contamination substantially, it is difficult to remove them all. To achieve the surface quality required for many flexible electronic applications, a planarising coating is necessary. This coating covers surface defects to enable a consistent, reproducible surface at any given point across the surface. The planariser coating comprises a low viscosity liquid that flows easily over film imperfections to create a smooth surface. Carrying out this process in a cleanroom ensures that the surface has low intrinsic and extrinsic contamination. Importantly, the planariser coating also has anti-scratch properties preventing subsequent scratching of the film during processing and handling. Therefore, thin display overlayers, typically tens of nanometers thick, are deposited with better quality. The choice of chemistry and coating thickness of the planarising coating has an impact on the physical properties of the planarised film. Factors to consider include maximum processing temperature during film processing and the chemistry of layers that will be deposited on the planariser surface. Suitable chemistry types that offer pencil hardnesses greater than 2H

range from predominantly inorganic materials, for example sol-gel derived coatings based on siloxane chemistry [8] to organic materials, for example polyfunctional acrylate coatings [9]. The surface quality can be defined in two ways, the intrinsic surface smoothness and the surface cleanliness. The intrinsic surface smoothness is a function of both polymer film type and film manufacturing route. The surface smoothness is captured typically over tens of micron square dimensions and is reported as the surface micro-roughness. Typical values range from  $R_a = 0.6\text{nm}$  for clear PEN film to  $R_a = 100\text{nm}$  for filled PET. The surface cleanliness is a function of the amount of external contamination on the surface and damage to the film, such as scratches. The former can be typically 40 microns long by 10 microns high with scratches typically often 150 micron long by 0.5 micron high. Both of these have sufficient height projection to cause a problem to a thin display layer.



**Illustration 1. Scratch on PET film surface**

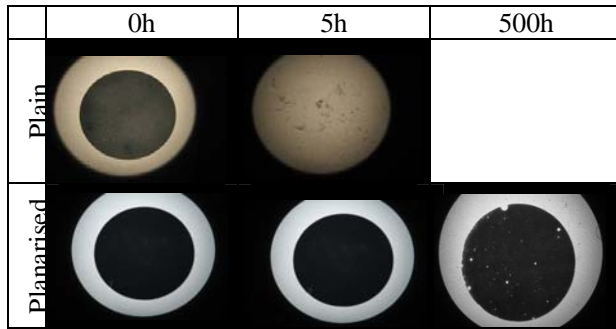
Experimental data in figure 1 shows peaks greater than 40nm on plain PEN (not planarised) surface and a planarised PEN surface over a 5cm by 5cm area. There are 10 peaks with height of 130nm on the planariser surface compared with 50 on the raw surface. The outer lying points, which are the most serious for proper display function, are completely removed in the case of the planariser.



**Fig. 1: White light interferometry plot over 5x5cm area of PEN plain surface and planarised PEN Film**

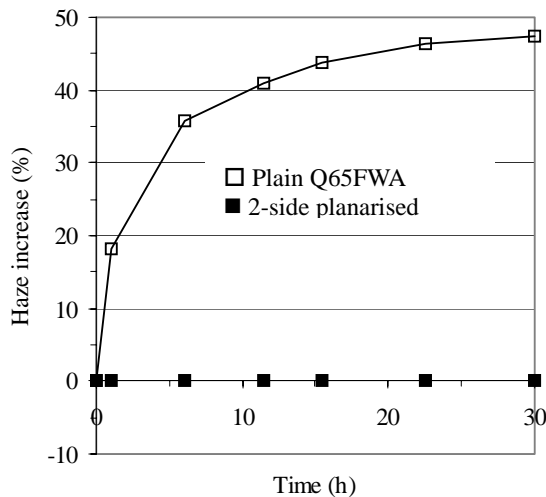
To quantify the beneficial effect of planarisation upon extrinsic debris and scratches is more difficult, as these features depend on sample handling and local environment of the film. Extrinsic debris, for example, may form in localised regions across the surface. By covering these features, the planariser surface ensures a consistent high quality robust surface, which does not vary from sample to sample. This is particularly important in flexible electronics, as one scratch in a region can interrupt a whole row or column of pixels in the display, even if the other areas are acceptable.

A practical example of the benefits of planarisation can be seen in table 1 when a thin barrier layer is applied on top of planarised and raw PEN film. To measure the substrate influence on quality of the barrier layer, a thin layer of moisture-sensitive calcium is deposited. The calcium is corroded after 5 hours when the barrier is placed directly on the PEN's raw surface. This can be partly explained by the non-removable extrinsic and intrinsic contamination surface projections causing pinholes in the barrier layer. These are largely absent in the planarised case reflecting smoother intrinsic surface and a reduction in extrinsic debris. The calcium area is largely intact at 500 hours of accelerated aging.



**Table 1: Effect of planariser layer on calcium degradation under accelerated aging (60°C, 90% r.h.)**

PET and PEN film contain 1.1wt% and 0.3wt% cyclic oligomer respectively. The presence of these on the surface is undesirable, as they can interfere with the TFT manufacturing process. The oligomer crystals are soluble in methylethylketone (MEK) and may be removed by washing. Our data has shown that planarised films produce much less oligomer deposition on the surface at high temperatures. The oligomer migration can be conveniently monitored by the surface light scattering that they induce, once the polyhedral hexagonal oligomer crystals migrate to the surface. In figure 2, in the case of the two-side planarised PEN, the increase in surface haze with time is less than 1%, compared to more than 45% for uncoated PEN. This benefit is particularly relevant for display processing at high temperatures, such as for organic- or inorganic TFT backplanes.



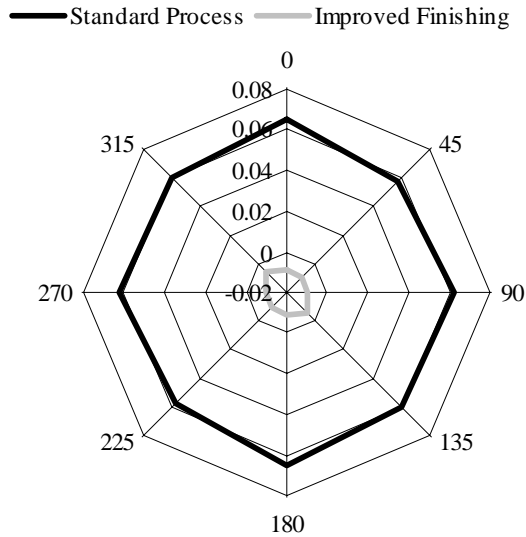
**Fig.2: The effect of planariser coating on oligomer surface migration at 200°C**

The scratch-resistance properties of planarised films have been measured by the Taber abrasion test. Results indicate a 50% increase in haze for uncoated PEN, compared to 1.4% in the case of the planarised surface. This ensures prevention of scratches and corresponding display problems on the next downstream process steps. In terms of optical properties, the planarised films offer good clarity and are suitable for top-emissive frontplanes like OLEDs. Planarised Teonex® Q65A film offers a 0.7% increase in total light transmission and a 0.3% decrease in haze, compared to uncoated PEN. This in part reflects the lower refractive index of the coating - in the range of 1.4 to 1.6 - compared to PEN (1.74). The tensile properties are largely unaffected by the presence of the planariser.

#### *Improved Finishing Process*

The thermal dimensional stability of a polymeric substrate for flexible electronics can vary widely depending on the choice of the polymer material. While amorphous polymer systems such as polyimide and polyethersulphone display excellent chemical inertness at elevated temperatures, they nevertheless suffer from poor thermal dimensional stability. They exhibit a relatively large coefficient of linear thermal expansion (CLTE) and an irreversible shrinkage from residual frozen-in strain, which can arise from the process history. PEN film possesses a lower average CLTE; due principally to its biaxial microstructure and it has been shown that with the appropriate process history, its thermal dimensional stability can be further improved [10-11]. This is described briefly in the following figures.

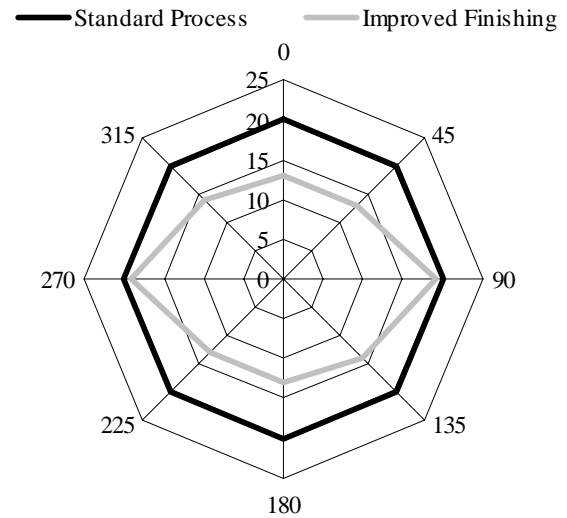
The thermal dimensional stability of a polymeric substrate will comprise two components, namely the irreversible strain recovery or shrinkage and the reversible expansion and contraction with thermal cycling. By careful control of the thermal finishing process of Teonex® film, it is virtually possible to remove the shrinkage behaviour of film. Figure 3 shows the shrinkage of Teonex® PEN film, after 30 minutes at 180°C and measured in four principal directions in the plane of the film. A sample selected from the centre of a wider film roll shows some anisotropic character as a result of the stretching ratios in the forward (MD, 0°) and transverse directions (TD, 90°) during manufacture. However, all values of shrinkage remain below 0.10%.



**Fig. 3: Shrinkage % (180°C, 30min) vs. direction in plane of Teonex® PEN film (0°=MD)**

In the figure, the same film, after the more controlled process treatment, is seen to retain the near isotropic behaviour. However, shrinkage is now essentially removed. In fact, the recorded values of around -0.01% after 30 minutes at 180°C indicate minor expansion, but these are close to the sensitivity of the measurement. The second factor which contributes to the dimensional stability, the CLTE, also shows a contrast between film with standard and with more controlled finishing stages to the manufacture.

Figure 4 illustrates that over a temperature range from 20 to 100°C, the CLTE, averaged in all directions, is seen to improve by around 25%, from a value of 20 ppm/K to 15 ppm/K. Again, the film is virtually isotropic in its plane. This improvement in thermal dimensional stability now offers more tolerance to fabrication processes where temperature cycles traditionally create issues around layer-on-layer registration.



**Fig. 4: CLTE (ppm/K) vs. direction in plane of Teonex® PEN film (0°=MD)**

### 3. Summary

There are a large number of demands on the plastic substrates in order to meet the requirements of flexible electronics. In this paper, we have shown that Teonex® Q65 and Melinex® ST506/504 are polyester substrates which can be used in flexible electronics. Planarised layers of coatings, when applied to these substrates, can enhance their surface smoothness and cleanliness, raise the surface hardness and retain the optical properties under thermal treatments that are typically used in display manufacturing processes. An improved finishing processed can be applied to Teonex® Q65 films. The results suggest that this process can further reduce the thermal shrinkage at high temperature and lower the CLTE of the film.

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