# The Impact of Thermal Stress, Mechanical Stress and Environment on Dimensional Reproducibility of Polyester Film during Flexible Electronics Processing

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## Abstract

*DuPont Teijin Films™ (DTF) have developed engineered substrates specifically for the flexible electronics market. Teonex® Q65, Melinex® ST506 and ST504 are biaxially oriented crystalline polyesters with the option of planarised surfaces. These films are emerging as competitive materials for the base substrate in OLED displays and active matrix backplanes. Given the demanding dimensional reproducibility requirements in the display applications, it is critical to control the several factors that can influence the film distortion in order to achieve the ultimate performance. This paper will discuss the impact of thermal stress, mechanical stress and the processing environment on dimensional reproducibility of polyester film and give examples of how this impacts on the film in device manufacture.*

## 1. Objectives and Background

There is currently considerable interest in flexible displays and many electronic based companies are actively researching flexible displays based on liquid crystal displays (LCD), organic light emitting diodes (OLEDs) and electrophoretic displays. To replace glass however, a plastic substrate needs to be able to offer similar properties in terms of clarity, dimensional stability, thermal stability, barrier, solvent resistance, low coefficient of thermal expansion coupled with a smooth surface. DTF have two families of polymers based on PET (Melinex®) and PEN (Teonex®). To achieve the dimensional stability required for display manufacture the film is commonly off-line stabilized in a process where the internal strain in the film is relaxed by exposure to high temperature whilst under minimum line tension [1-7]. Teonex®Q65FA is a one side pretreated, heat stabilized PEN film. Since PEN is a higher temperature performance substrate, Teonex®Q65FA emerges as the leading base substrate for OLED displays and active matrix backplanes. Dimensional reproducibility is a key property of the base substrate and, in addition to temperature cycling, the mismatch in properties between organic and inorganic layers can lead to further distortion issues. Several factors, which include thermal, mechanical and environmental properties, influence these key parameters and these must all be controlled to ensure that the ultimate in dimensional reproducibility is attained.

## 2. Results

### 2.1 The Effect of Thermal Stress on Dimensional Reproducibility

Teonex® and Melinex® are produced using a sequential biaxial stretching technology, which is widely used for many semi-crystalline, thermoplastics [8]. The process involves stretching film in machine and transverse directions (MD and TD) and heat setting at elevated temperature. As a consequence, a complex semi-crystalline microstructure develops in the material, which exhibits remarkable strength, stiffness and thermal stability [9,10]. The film comprises a mosaic of crystallites or aggregated crystallites accounting for nearly 50% of its weight and which tend to align along the directions of stretch. Adjacent crystallites may not however share similar orientations. Crystallites show only a small irreversible response to temperature, which may take the form of growth or perfection. The non-crystalline region also possesses some preferred molecular orientation, which is a consequence of its connectivity to the crystalline phase. Importantly, the molecular chains residing in the non-crystalline region are on average slightly extended and therefore do not exist in their equilibrium Gaussian conformation.

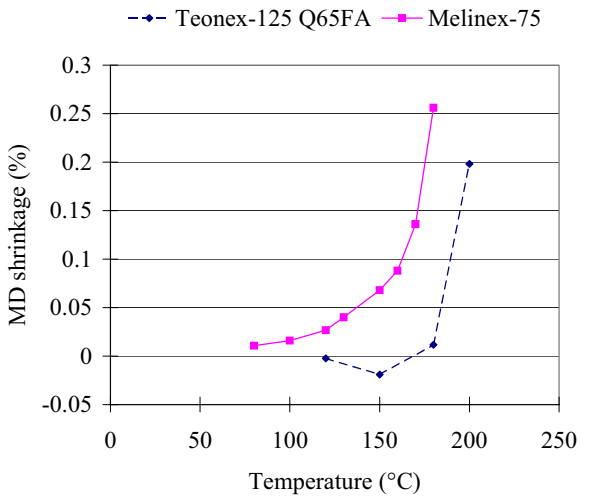


Figure 1: Shrinkage of heat stabilised PET and PEN film versus temperature

Teonex® and Melinex® films can be further exposed to a thermal relaxation process in which the film is transported relatively unconstrained through an additional heating zone. Some additional shrinkage is seen which signifies a relaxation of the molecular orientation in the material, which is believed to occur exclusively in the non-crystalline regions [11].

Figure 1 illustrates how the shrinkage of heat stabilised Melinex® and Teonex® changes with temperature. This clearly shows the higher temperature performance of Teonex® relative to Melinex®. For applications requiring low shrinkage above 150C, Teonex® Q65 is the preferred option.

It is generally believed that the irreversible thermal shrinkage of semicrystalline biaxial film is dictated by the state of the non-crystalline phase in the material. As well as experimental evidence, the success of phenomenological models also justifies this conclusion. These models can assume a simple elastic or spring-like behaviour for the non-crystalline component of films. By taking account only of initial and final strain in film during processing and associated temperature, models can predict the shrinkage recovered when reheating to any temperature.

Figure 2 shows the shrinkage of 200C heat stabilised Teonex® Q65 film, measured at 180C after 30 minutes. Although performed on sheet samples in the laboratory, an equivalent line tension was applied during treatment and so final values are plotted as a function of "line tension" which would be used during a real stabilisation process.

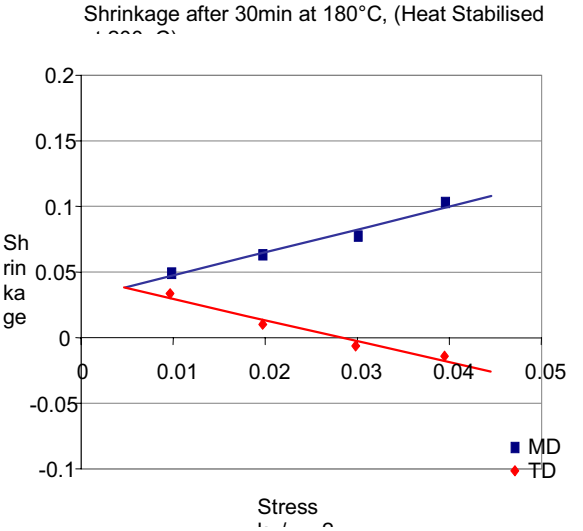


Figure 2: Shrinkage of heat stabilised Teonex®Q65

This behaviour is typical for biaxial polyester film and can be accurately predicted using the model outlined above. The results are consistent with the fact that the shrinkage behaviour of PEN film is related entirely to the condition of the non-crystalline regions of the material, and that efforts to reduce or eliminate this instability must focus on re-establishing the equilibrium conformation of the chains which exist in the disordered state.

### 2.2 The Effect of Mechanical Stress on Dimensional Reproducibility

Figure 2 clearly shows a dependence of the thermal shrinkage on the tension applied to the PEN film during its final stabilization treatment. Shrinkage in both machine and transverse directions are shown and while "zero shrinkage" is measured in the TD, this performance cannot be achieved simultaneously in the MD. The explanation is that while the film is free to relax without constraint in the TD, the film will always experience an applied tension in the machine direction, in order to transport it through the stabilizing stage. In theory, zero tension will permit full relaxation of the film in that direction and remove all strain memory. This is unlikely in practice for two reasons:

1) Roll to roll processing requires a finite line tension

2) The modulus of PET and PEN at relaxation temperatures is low

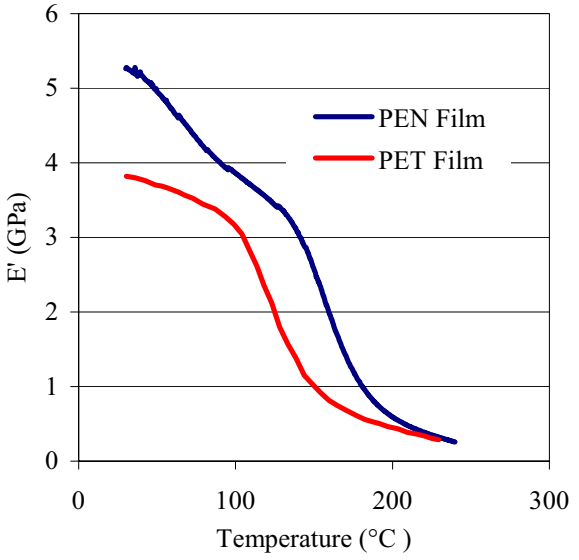


Figure 3: Stiffness of polyester films versus temperature

As depicted in figure 1, the storage modulus is decreasing from 3.5 GPa at Tg to less than 1 GPa at temperatures above 180C for Teonex® but 150C for Melinex®, the conventional temperature regime for each film type stabilisation. Any small tension or load applied to film at that temperature will therefore impose strain in the material, which will be "frozen–in" upon cooling and reappear as shrinkage upon reheating. The implication of this is that if the film is constrained through a thermal processing cycle e.g. in a roll-to-roll process or by lamination to a rigid carrier, the strain introduced into the film may result in subsequent shrinkage on reheating.

| Teonex® thickness (microns) | Rigidity ($N \cdot m/10^{-4}$) | Rigidity relative to 125 micron film |
|---|---|---|
| 125 | 15 | 140 |
| 175 | 40 | 390 |
| 200 | 60 | 580 |

Table 1 Effect of thickness on Teonex® rigidity

Measures of stiffness such as the Young's Modulus are thickness independent and do not indicate how rigidity will change with thickness. Rigidity (D) can be defined by $D = Et^3/12(1-\nu)$, where E is the elastic modulus (6.1GPa), t the thickness and ν the Poisson ratio (0.33) [5] It can be seen from table 1 that the thickness of the film has a significant effect on the rigidity of the film. A 200 micron PEN film is 4 times more rigid than 125 micron PEN.


Figure 4. Distortion through a-Si deposition (slide courtesy of Flexible Display Centre, ASU)

This extra rigidity can be exploited to reduce the stress and resulting distortion as evidenced by the difference in distortion measured through a-Si TFT process on 125µm Teonex® compared to 200µm Teonex® Q65. For the latter, the distortion through the process is reduced from ca 175ppm to 100 ppm (Fig. 4).

### 2.3 The Effect of Environmental Conditions on Dimensional Reproducibility

It has been shown in a previous publication [12] that a film expands by approx. 45ppm in the three dimensions for every 100ppm moisture absorbed. Knowing the solubility and diffusion rate of moisture in PEN as a function of temperature, it is possible to model the impact of various environmental conditions on moisture content changes and hence volumetric changes in the different thicknesses of film.

At room temperature, initially dry films can typically take over 12 hours to reach moisture equilibrium. For instance, at a processing temperature of 100°C, the film may take a longer time to reach its equilibrium moisture level than the time necessary to carry out the processing step. Ideally, the film should be allowed to equilibrate before any registration points are established.

### 2.4 Cyclic Oligomers

PET film contains 1.1wt% cyclic oligomer which can migrate to the surface, if the film is held at elevated temperatures for tens of minutes. The

presence of these oligomers on the surface can interfere with the TFT manufacturing process. Teonex®, at 0.3wt%, has significantly lower cyclic oligomer content, when compared with Melinex® and there is significantly less migration to the surface of the film (fig. 5). The cyclic oligomer crystals form on the surface and can be removed by washing the film with MEK (methylethylketone) solvent. Shortening the time the film is held at temperature and reducing processing temperatures can minimise cyclic oligomer migration. Where this is not possible, changing to Teonex®Q65 offers an alternative option to minimizing the presence of cyclics on the surface.

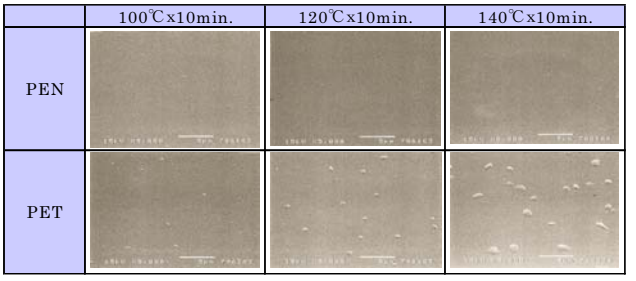


Figure 5: Cyclic oligomer growth on the surface of PET compared to PEN

### 2.5 Examples of Film in Use

As shown in fig. 4, The Flexible Display Centre at Arizona State University have shown that distortion of Teonex®Q65 can be reduced to 100ppm at processing temperatures up to 180C [13] during the fabrication of a-Si TFT backplanes. PARC have also reported distortion of <100ppm via low temperature PECVD [14] during a-Si TFT backplane preparation. In both cases, the control of thermal stress, mechanical stress (a thicker film) and the environment (moisture) have been instrumental in achieving this performance.

## 3. Impact

As discussed above, excellent dimensional stability of the order of 100ppm at processing temperatures of 150C can be achieved with Teonex®Q65 provided that (i) thermal stress i.e. shrinkage via off-line stabilisation, (ii) mechanical stress i.e. film thickness and tension during processing and (iii) environmental conditions are controlled. Teonex®Q65 also has less migration of cyclic oligomers to the film surface at elevated temperatures. The evidence from device manufacturers indicates that through careful control of processing, device manufacture of a-Si TFT backplanes on Teonex® Q65 is viable.

## 4. References

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