The role of the thermoplastic matrix in forming processes of composite materials.

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Abstract.

The application of continuous fibre reinforced thermoplastics in load bearing primary constructions is still limited. To increase the use of these composites, thermoforming processes must be developed. This paper considers the investigation of the influences of the presence of thermoplastic matrix during deformation of laminates in a thermoforming process.
In the first part the investigations are focused on rubber forming as a promising fast manufacturing method. In all thermoforming processes however, the thermoplastic matrix will act as a lubricant inside the deforming fabric. Intraply shearing and interply slipping are two laminate deformations where this lubrication has an important influence. Some deformation tests and their results will be described in this paper.
Another forming process for fibre reinforced thermoplastics is diaphragm forming. During this process the laminate is placed between two thermoplastic foils. The processing parameters pressure, temperature and time influence both the deformation of the laminate and the deformation of the foils. This influence is different for the flat laminate and the foils due to their different mechanical and thermal properties. It is therefore necessary to investigate a suitable compromise in the processing window.

Introduction.

Thermoplastic composites have several advantages over thermoset matrix composites and metals which makes them very interesting for the use in aircraft structures. It is therefore surprising that despite the advantages like high speed manufacturing, there is still a reserved use of these materials in commercial manufacturing industries in general and in aerospace industries in particular. The reason for this reserved use is the lack of knowledge of the processing of these materials. It is therefore necessary to develop a good understanding of the processing science so that the combinations of high speed manufacturing and high material properties will be realized for the particular composite parts to be made.

The University of Technology Delft supports both the material producers and the composite parts manufacturers with manufacturing and processing technology. Projects on thermoplastic composites are integrated in Ph.D. investigations and contract work. In this paper we will focus on the (sub)processes that take place during the thermoforming of fabric reinforced thermoplastics as their optimisation will be the key to a successful production of composite parts.

Two basic thermoforming techniques that are investigated at TU Delft, diaphragm forming and rubber forming, will be used to illustrate the following processes. First the deformation of the fabric reinforcement during the thermoforming of composites will be discussed. Secondly the role of the matrix viscosity and foil viscosity in the forming processes will be discussed.

Rubber forming and diaphragm forming.

A unique property of thermoplastic composites is their thermoformability. A fibre reinforced thermoplastic sheet is formable into a two- or three- dimensional product by heating it to a temperature well above its glass transition temperature and by applying pressure. This property makes it possible to achieve a relatively low cycle time for the production of thermoplastic composite parts. The cycle time is determined by the time necessary for the heating, (de)forming and cooling of the composite material. The thermoforming techniques are distinguished by their different forming devices.

In rubber forming the heated flat laminate is positioned between two dies (fig.1). The dies are fixed in a fast closing press and the laminate is shaped into a two or three dimensional product by these two dies. Essential is the fact that one of the dies is (partly) made of rubber whereas the other is a rigid one, mostly made of metal. The form of the dies provides the needed local force to deform the laminate. The flexibility of the rubber can lead to a homogeneous pressure distribution on the composite product that is consolidating in the mould. Mostly the positive die matches with the negative one, but that is not necessarily true. Sometimes it is even favourable to under- or overdimension the rubber die in order to avoid or create a certain local sheet pressure and make a certain product(form) possible. In this way it is not a matched die process. The technique is called rubber forming to distinguish this thermoforming technique from rubber pressing, which uses one rigid mould and a flat rubber pad.

Because of the necessity of the reinforcing fabric in the composite sheet to adjust to the mould, forces must be introduced to deform the fabric. This can be realized by using clamping facilities and special die formes. On the other hand, local forming pressures on the sheet must not exceed a certain critical level to avoid excessive matrix flow. When these subjects are understood and well introduced in the mould design of the rubber forming technique, low cycle times (2-5 minutes) can be obtained and high speed manufacturing oawthermoplastic composite parts is a fact.

In diaphragm forming the laminate is placed between two diaphragms (fig.2). The diaphragms are clamped onto the mould after which vacuum is applied between the diaphragms. The mould is then placed into an autoclave and heated to the processing temperature. After reaching the processing time, an hydrostatic pressure will be created which forces the laminate into the mould. The laminate is not clamped and is allowed to slide within the diaphragms. This sliding action creates tensile stresses in the sheet from interfacial shear stresses that reduces wrinkling [*ref.1*]. The diaphragms have therefore the function of blankholder. Diaphragm forming gives products with an excellent quality. The cycle times are, however, longer than the cycle times for rubber forming.

Fabric deformation.

The deforming capabilities of a composite sheet is limited by the deformability of the reinforcing fabric. Fabric deformation therefore is a necessary condition during the thermoforming of fabric reinforced composites [*ref.2*]. There are two main deformations that have to take place inside a laminate to realize a three dimensional product, interply slip and intraply shear.

Interply slip is slipping of the different layers relative to each other when the laminate is bent (fig.3) [*ref.3*]. The phenomenon must already take place in two dimensional deforming of a laminate and therefore it is important that this slipping of the fibre layers is facilitated by decreasing friction between the layers.
In particular when using the rubber forming technique, high local (normal) forces on the laminate occur during forming. The normal forces increase the friction between the layers at the point of application. Interply slip can be obstructed here. On the other hand, areas of the laminate exist where in the forming phase normal force is absent. Fabric deformations are easily allowed there.
It is obvious that the liquid thermoplastic matrix plays an important role in this process as a lubricant. This will be described in the next section.

Another deformation in case of fabric reinforcement is intraply shearing or Trellis effect [*ref.4*]. The term 'intraply shearing' indicates that this fabric deformation occurs inside one ply. To accomplish a three dimensional form out of a flat laminate, the interlacing fibre bundles of the reinforcing fabric will turn and the crossover angle of the bundles will change as depicted in figure 4. In general a minimal crossover angle can be determined that can be obtained within a deformation process. In figure 5 some maximum shearing angles are given of matrix-less glass fabrics, determined according to the method of Behre [*ref.5*].
During intraply shearing, the deformation can be obstructed by two kinds of friction. The first one is the regular friction of the rotating fibre bundles moving with regard of each other. The second kind of friction occurs at the crossover points of the interlacing bundles. It obstructs the rotation of the fibre bundles.

Matrix lubrication.

Theoretically, the influence of the presence of thermoplastic resin can be twofold. First the viscous matrix acts as a lubricant and decreases the internal friction during the motion of fibrebundles in a laminate. Thermoforming will be facilitated in this way and lower forming forces on the laminate are possible. Secondly, the high viscosity of thermoplastic matrices can lead to obstruction of the motion of fibrebundles in a laminate. Visco-elastic effects can occur when fabric deformations take place too quickly.

In practice, both influences will be present in a thermoforming process. In fast forming processes like rubber forming, the visco-elastic obstruction will be of more importance than in slower processes like diaphragm forming. Lubrication of the fabric deformations will be affected by several variables of the composite material and the forming process. A few aspects of the influence of matrix will be dealt with in this section.

When a fabric reinforced laminate is bent during a thermoforming process, stresses are created in each fabric layer until a certain shear yield stress is exceeded in the resin rich layers between the fabric layers and interply slip starts [*ref.6*]. Experiments are being carried out on glassfabric reinforced PEI to determine the interply slip behaviour at thermoforming temperatures. In the experiments, schematically shown in figure 6, one layer is pulled out of the heated and presurized laminate.
Figure 7 gives the results for a five layer laminate. It is shown how the shear yield stress τ_o (which determines the conditions where forming is possible) decreases when thermoforming temperature rises (lower matrix viscosity). This is especially true when a higher normal stress is applied to the surface of the laminate. An increase of the normal stress on the laminate of 0.7 MPa nearly doubles the shear yield stress. It is therefore important to minimize laminate surface forces that are necessary with rubber forming.
Another possibility to decrease the initial shear stress is shown in figure 8. By increasing the resin rich interlayer, τ_o strongly decreases. The amount of thermoplastic matrix that can act as a lubricant determines the point at which interply slipping will start.

Increasing forming temperature and decreasing (local) normal pressure will lower the necessary forming forces during a thermoforming process. This is not only the case for interply slip but also for intraply shear of the reinforcing fabric.
Some preliminary results, shown in figure 9, show the influence of forming temperature on the total shear deformation in time. It appears that the final total amount of shear is the same at both forming temperatures and therefore independent of matrix viscosity. However, the time in which this total deformation is realized is shortened by applying a higher thermoforming temperature. In a non-isotherm process, as rubberforming is, time plays an important role because of the instant cooling down of the laminate between the dies [*ref.7*]. The processes are still under investigation.

Matrix and diaphragm viscosity.

Glass and Carbon fabric reinforced PEI prepreg of Ten Cate and Upilex-R diaphragms of ICI were used for the diaphragm forming of hemispheres. DMTA analyses of PEI and Upilex-R were made for the determination of the glass transition temperature of PEI (Ultem) and PI (Upilex-R) respectively 215 °C and 265 °C (fig.10 and 11). The viscosity of the matrix has to be low because of the influence on the fabric deformation as mentioned in the previous section. The processing temperature has therefore to be well above the Tg of the matrix. The processing temperature has also to be well above the Tg of the diaphragms in order to obtain high elongations. On the other hand the viscosity of the foil has to be high in order to avoid fibre wrinkling. The maximum processing temperature of the Upilex-R diaphragms is 400 °C since Upilex degrades very quickly at temperatures above 400 °C. The processing temperature of the first test was therefore 400 °C.

The hemispheres made at 400 °C showed three phenomena namely: dry spots along the edge (fig.12), Upilex-wrinkling (fig.13) and fibre wrinkling (fig.14). The dry spots are caused by stripping of the matrix which indicates that the viscosity of the matrix is too low. Decreasing the processing temperature to 370 °C increases the viscosity of the matrix in such an extend that stripping of the matrix will not occur anymore. The Upilex- and fibre wrinkles indicate that the viscosity of the diaphragm is also too low. In order to increase the viscosity of the diaphragm the processing temperature was lowered to 300 °C which is still high enough to obtain the necessary elongation. The Upilex wrinkes in circumferential direction are disappeared but there are, however, still Upilex wrinkling in the four corners of the hemisphere and there are also fibre wrinkles (fig.15). Decreasing the processing temperature to an even lower temperature resulted in failure of the lower diaphragm.

At the University of Technology Delft an investigation on the impregnation of Upilex-R with solvents, like e.g. NMP, DMF, has just started. The goal of this investigation is to change the properties of the diaphragm in such a way that Upilex- and fibre wrinkling will not occur with diaphragm forming. Preliminary tests at a processing temperature of 300 °C show that there is indeed no Upilex- and fibre wrinkling. This means that the solvent increases the stiffness (viscosity) of the diaphragm while the maximum elongation remains the same or does not decrease dramatically. Investigation of the influence of the solvent at the mechanical properties of the foil is just started.

Conclusions.

* Due to its important lubrication function, the thermoplastic matrix decreases interply friction and therefore facilitates slip deformation during thermoforming processes.
* Increased (local) surface pressure on the laminate has an important negative influence on the formability of the laminate.
* Laminate deformation is facilitated at higher thermoforming temperatures, but the final total amount of intraply shear will be determined by the reinforcing fabric.
* In diaphragm forming, the viscosity of the matrix may not be too low due to the creation of dry spots along the edge of the hemisphere by stripping of the matrix.
* The viscosity of the diaphragm may not be too low in order to prevent diaphragm wrinkling and fibre wrinkling.
* Impregnation of Upilex-R with a solvent, like e.g. NMP, DMF, has a beneficial effect on the properties necessary for diaphragm forming.

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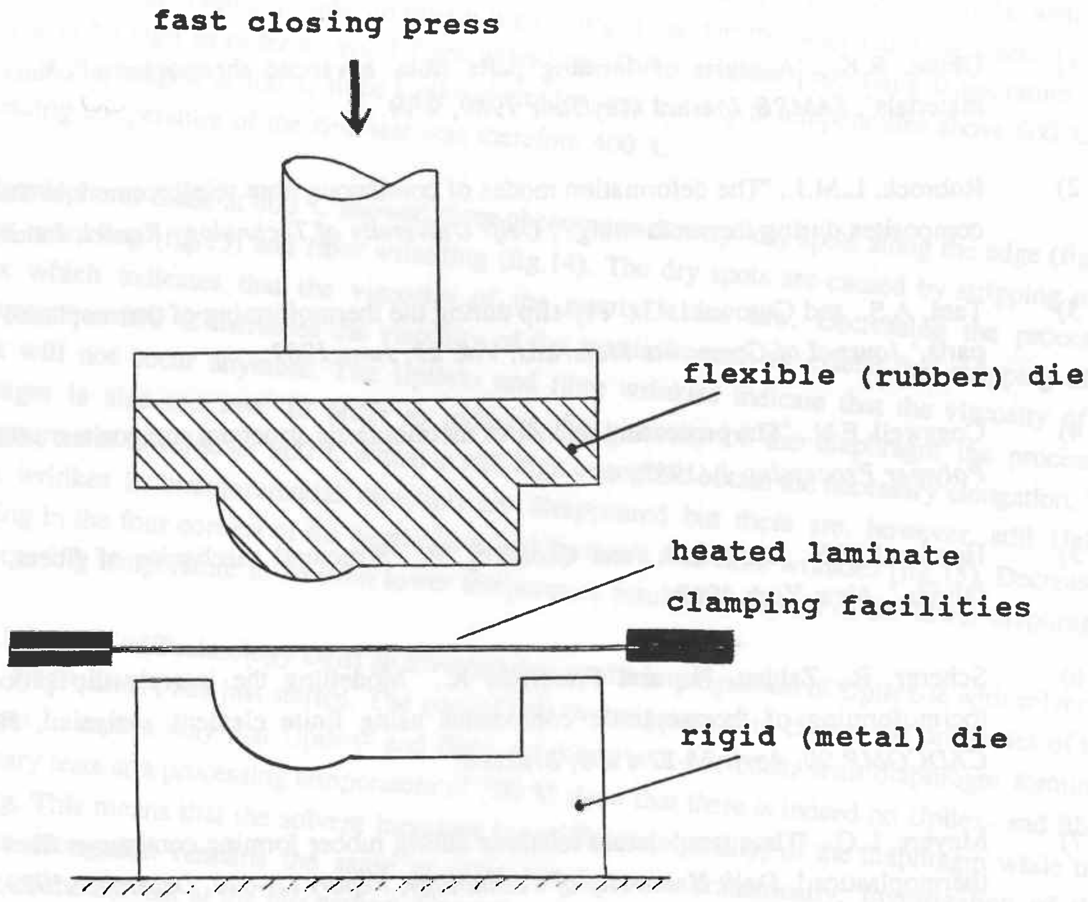


Figure 1.
Schematical view of the rubberforming technique.

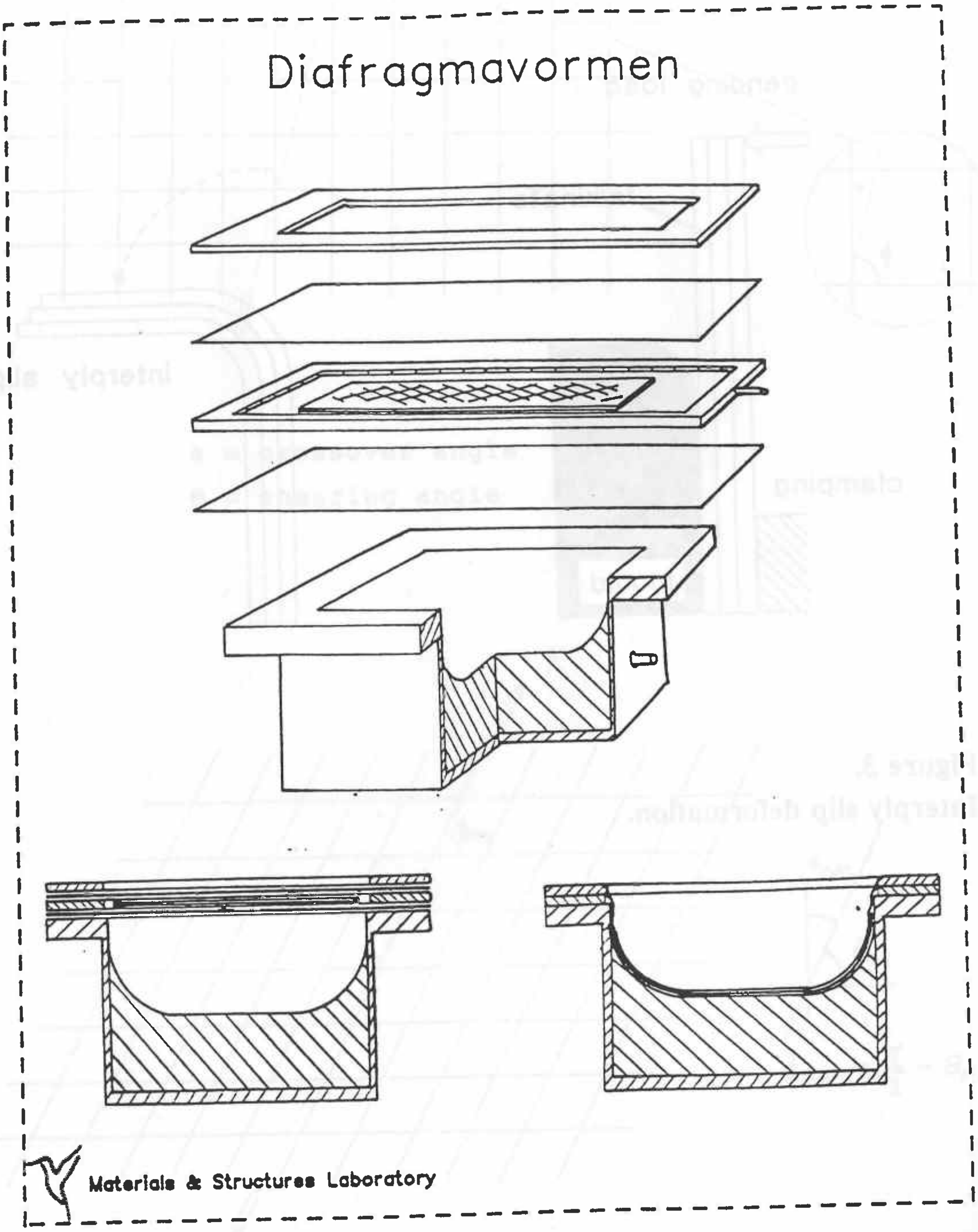


Figure 2.
Diaphragm forming.

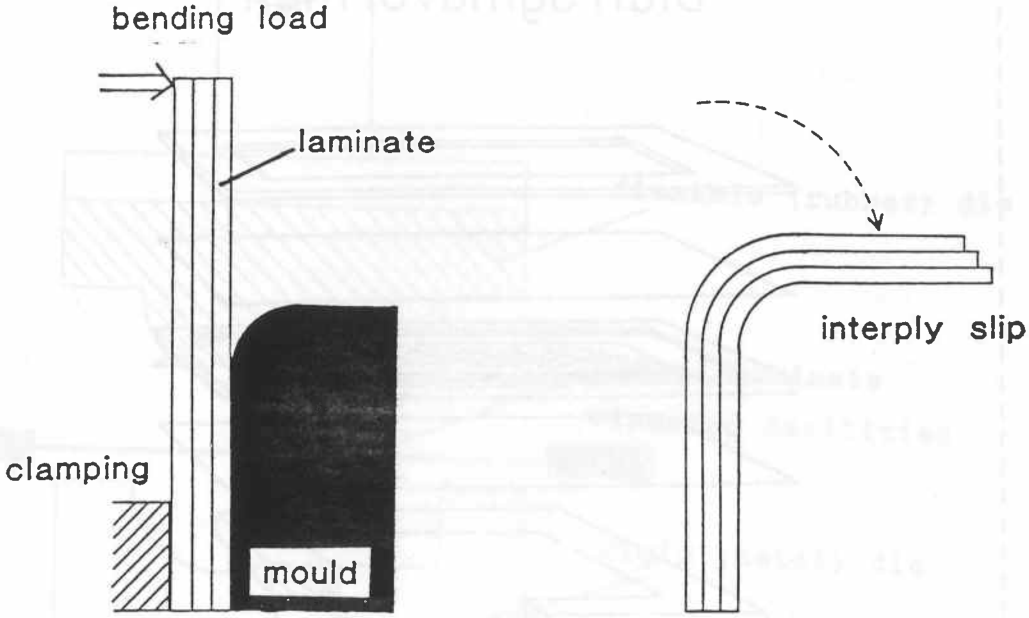


Figure 3.
Interply slip deformation.

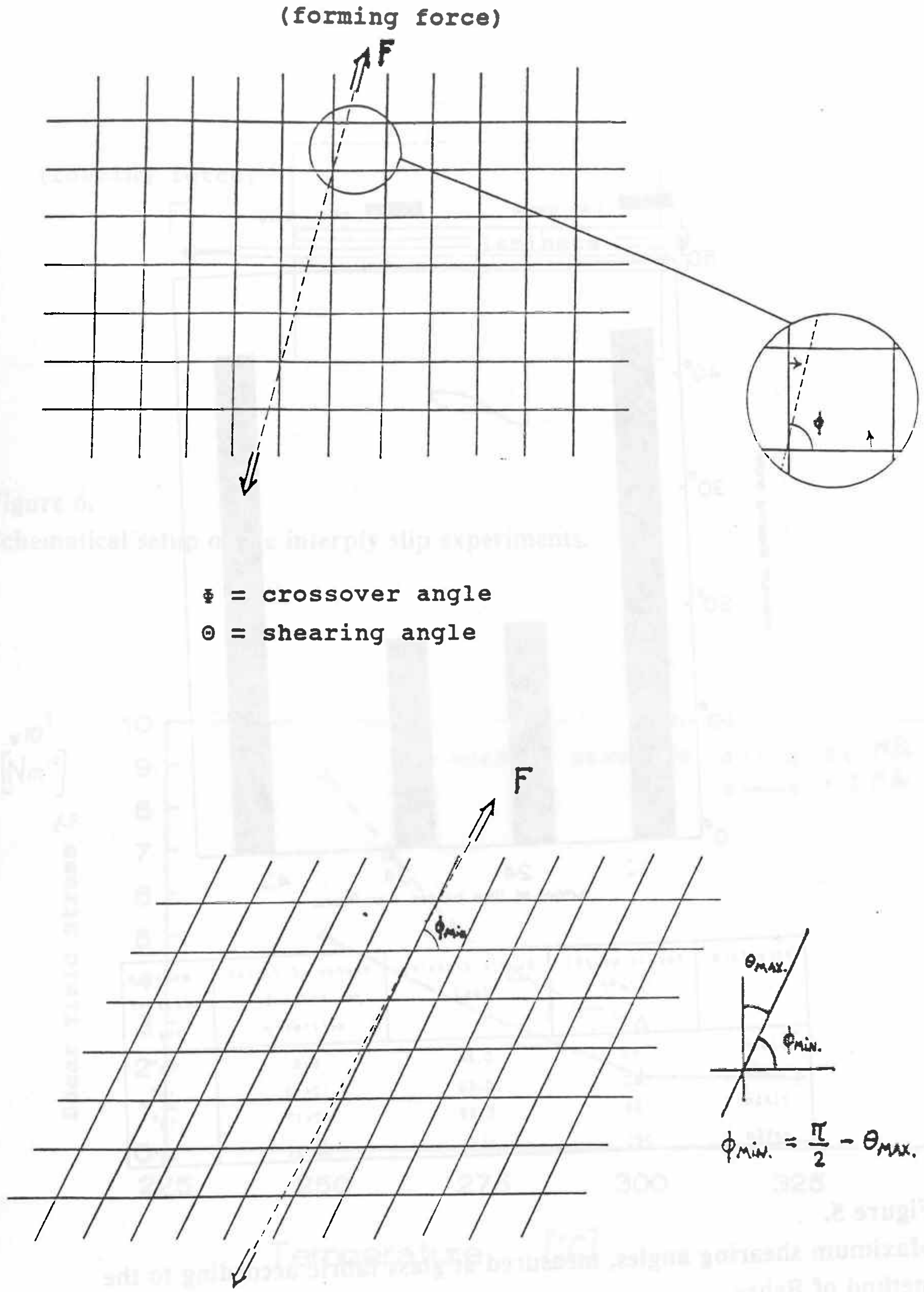
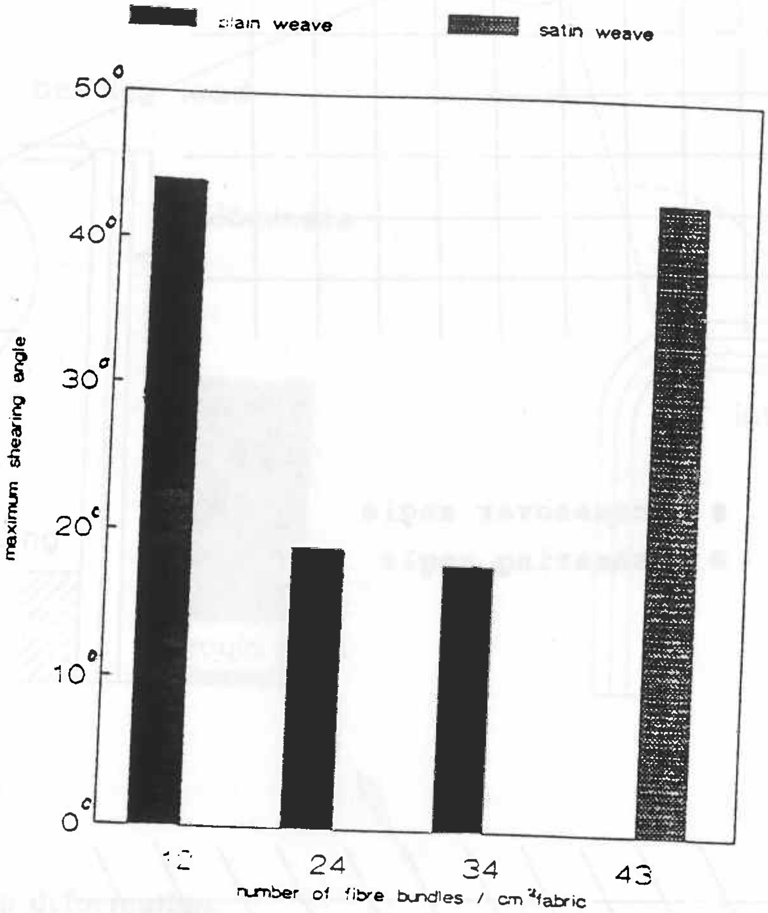


Figure 4.
Intraply shear deformation.



Weave form	fabric weight [g/m²]	bundle diameter [mm]	number of fibres per cm² fabric weft+warp	maximum shearing angle
plain	85	0.54	6+6	44°
plain	160	0.60	12+12	19°
plain	196	0.59	17+17	16°
satin	300	0.52	22+21	43°

Figure 5.
Maximum shearing angles, measured at glass fabric according to the method of Behre.

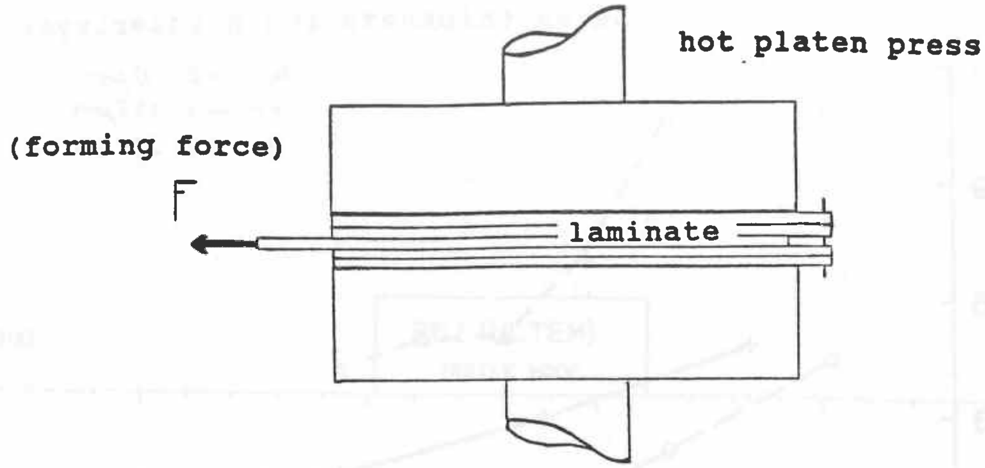


Figure 6.
Schematical setup of the interply slip experiments.

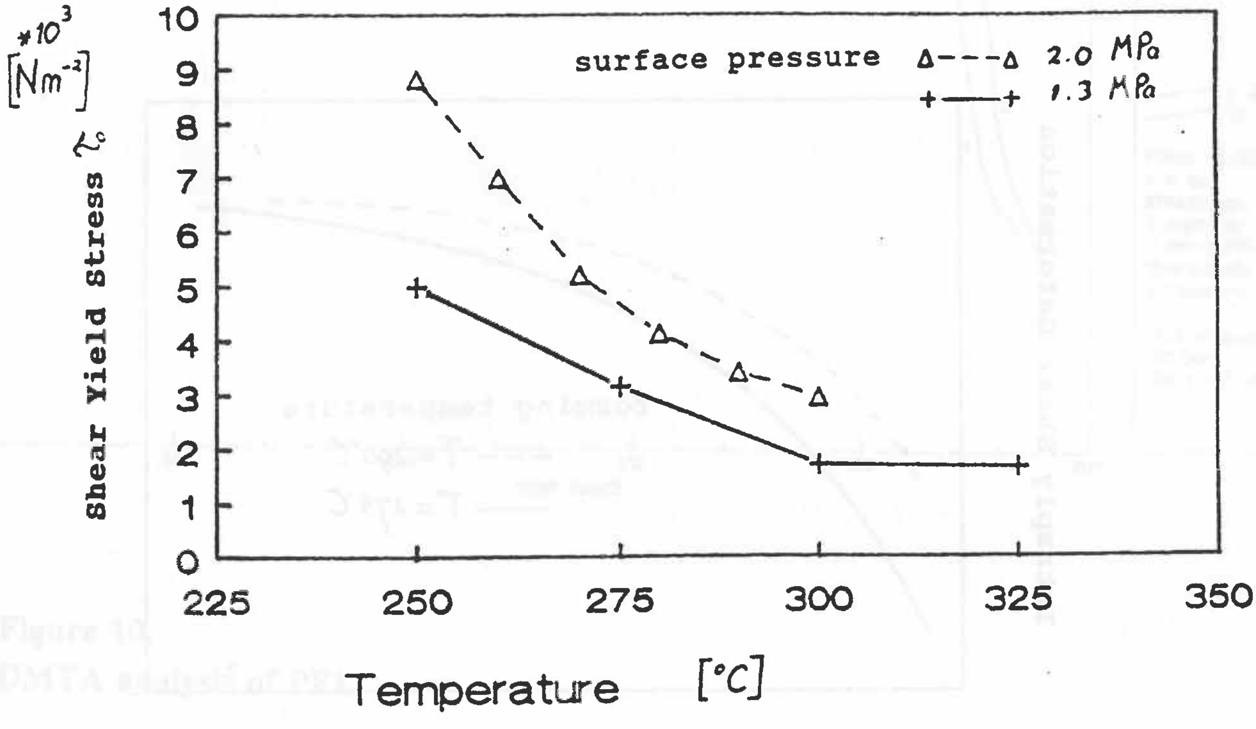


Figure 7.
Results of interply slip measurements of a 5 layer glassfabric/PEI laminate.

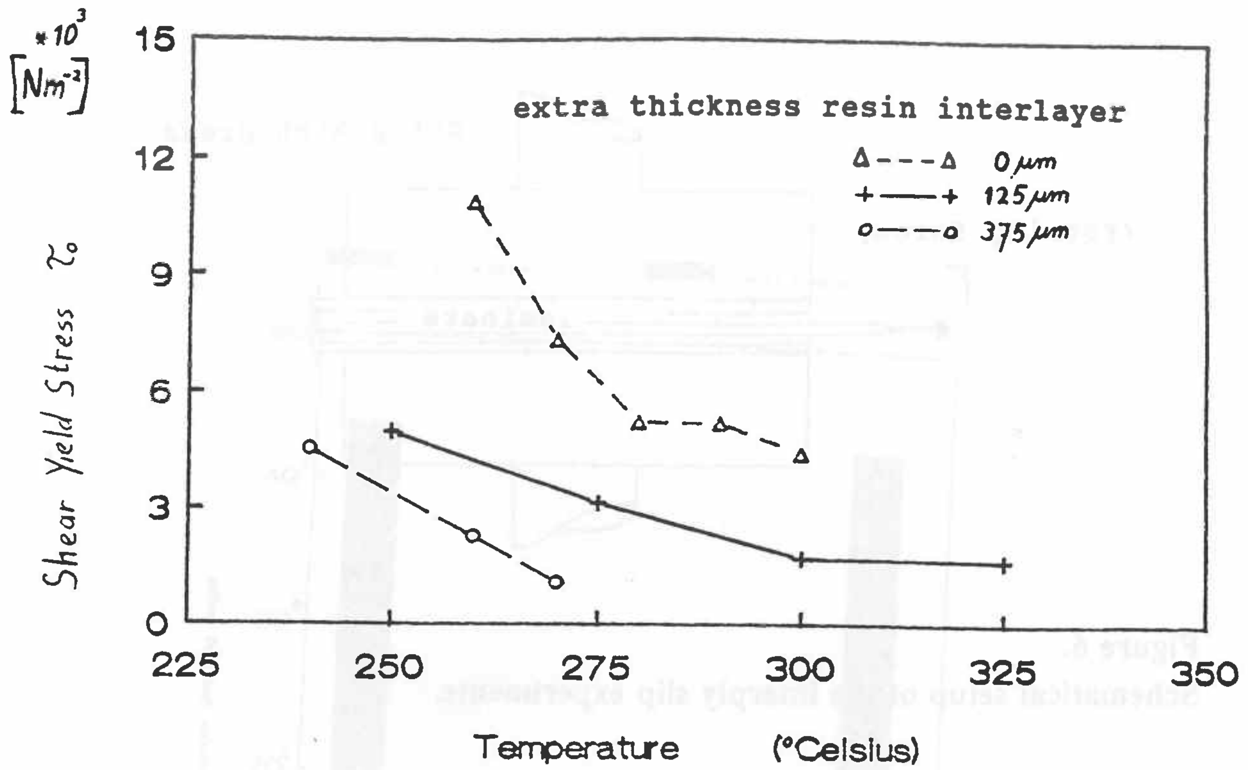


Figure 8.
Results of interply slip measurements of a 5 layer glassfabric/PEI laminate.

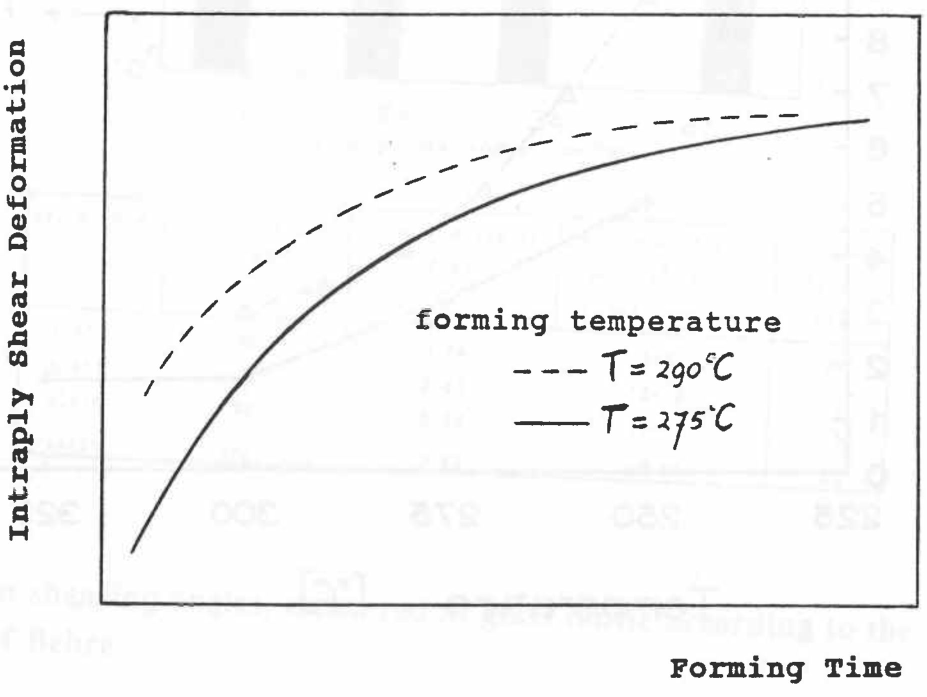


Figure 9.
Preliminary results of intraply shear deformation.

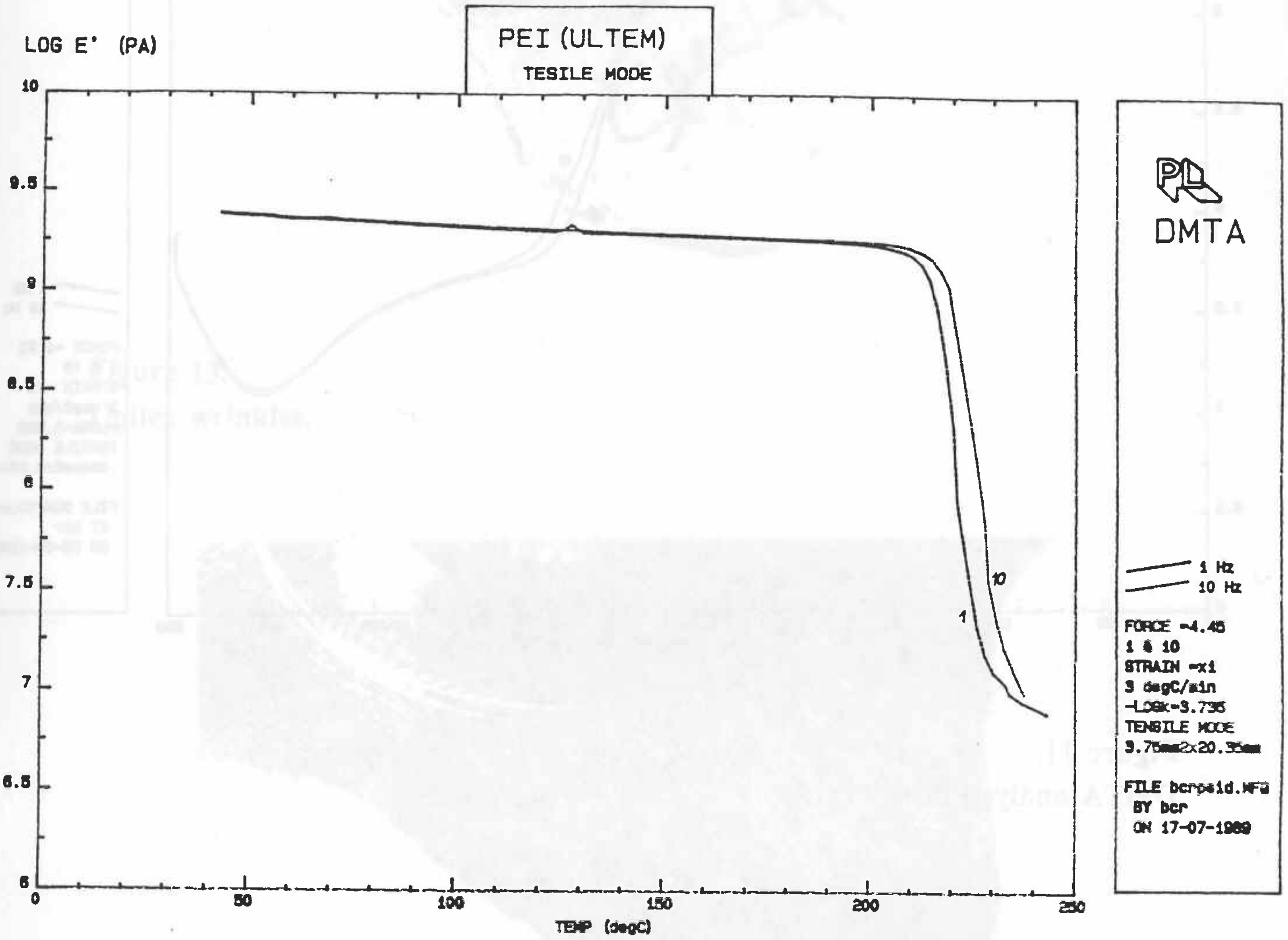


Figure 10.
DMTA analysis of PEI.

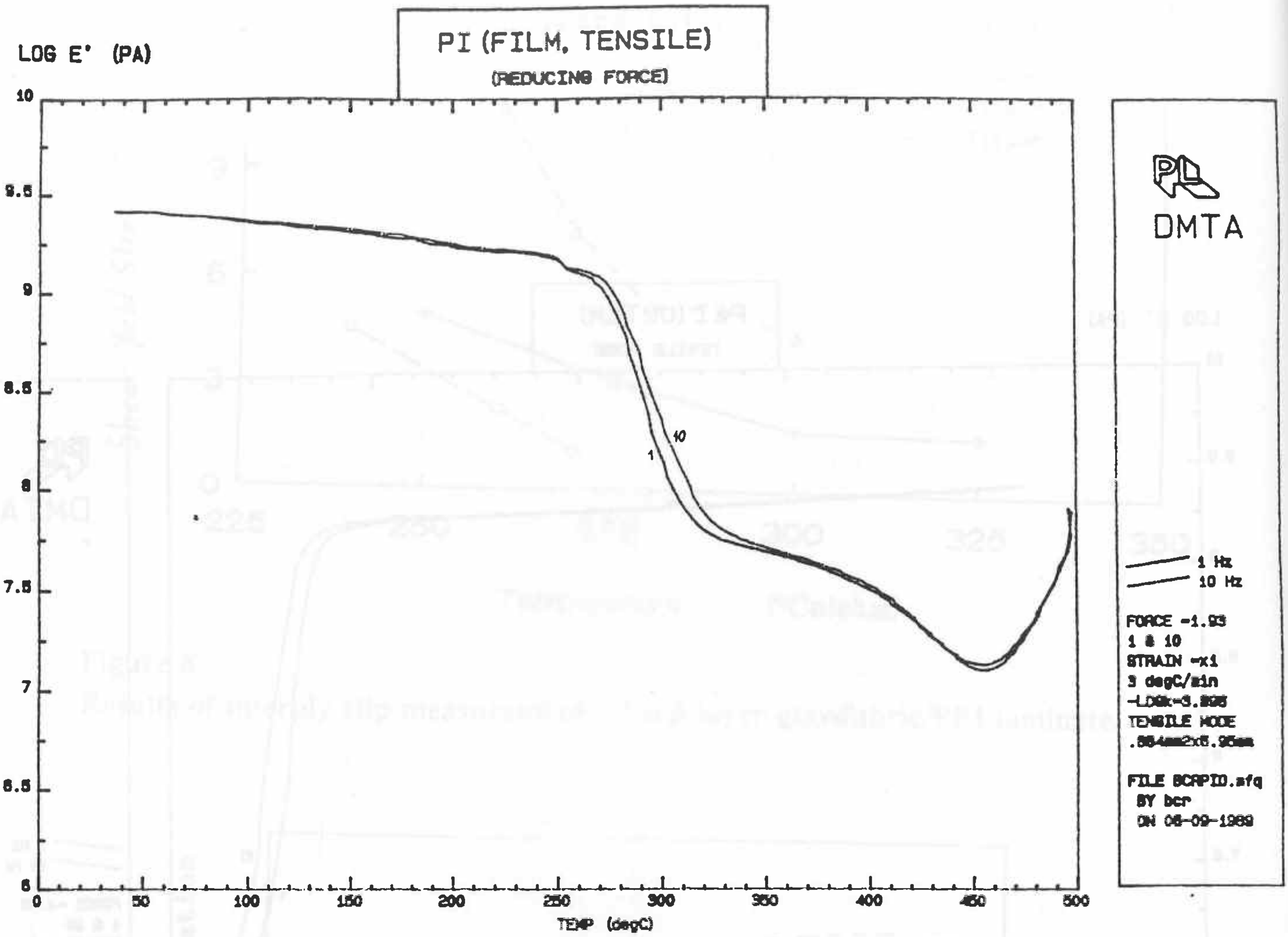


Figure 11.
DMTA analysis of Upilex-R.

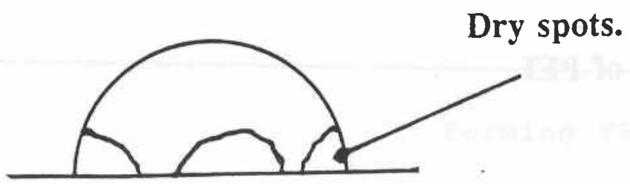


Figure 12.
Stripping of the matrix.

Figure 13.
Upilex wrinkles.

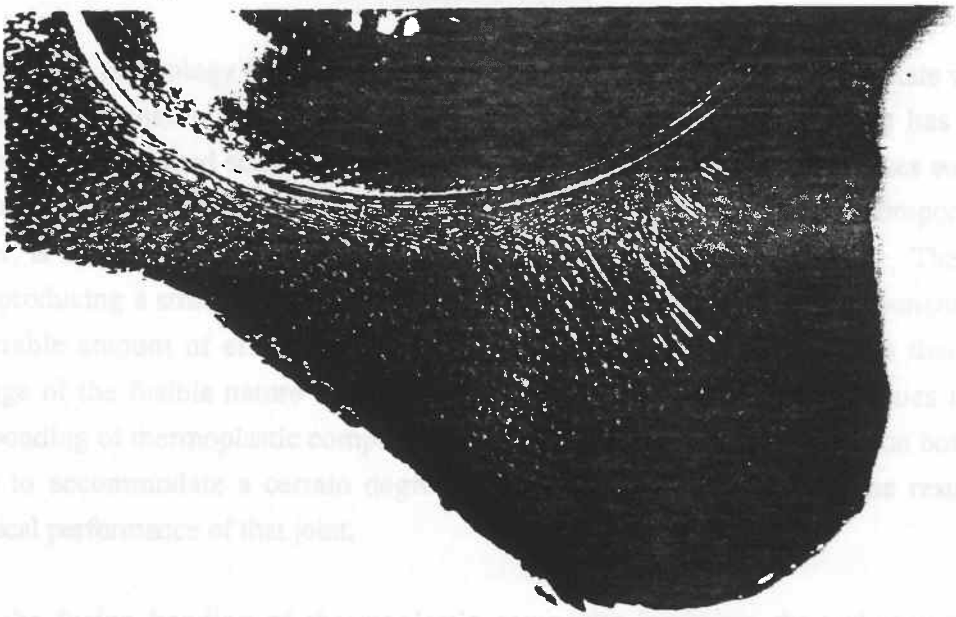


Figure 14.
Fibre wrinkles.