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# SERIAL SECTIONS THROUGH A CONTINUOUS FIBER-REINFORCED POLYMER COMPOSITE

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ABSTRACT

The microstructure of a unidirectional glass-fiber composite material is described seeking especially for the influence of the stitching perpendicular to the reinforcement. Serial cuts are performed through the composite and the microstructure is quantified using global parameters and linear morphological analysis. A key result is that the stitching induces variations in fibers spacing within the yarns and in the matrix volume between the yarns. This can affect noticeably the flow of the resin during the manufacturing process and also the mechanical properties of the composite.

Keywords: microstructure, serial sections, stitched composites, unidirectional fiber composites.

## INTRODUCTION

Continuous fiber-reinforced polymer composites are used in a wide range of applications, from aeronautics to sports (Berreur *et al.*, 2002). The purpose of the reinforcement is to offer specific mechanical solutions in terms of weight, performance and price. In most cases the reinforcement is made of glass or carbon fiber yarns arranged in bi-dimensional fabrics, 3D preforms or mixed structures such as stackings of multiply multiaxial fabrics (see Lundström, 2000 and Lomov *et al.,* 2003 for details). Owing to their versatility, bi-dimensional fabrics are commonly used. Among these fabrics, the woven ones are made of yarns orientated along two directions, the weft and the warp. Due to its simplicity, a stacking of unidirectional fabrics is the starting point for researches on liquid molding process (Gebart, 1992; Papathanasiou, 1996) and mechanical properties (Alberola *et al.*, 1999) of composites. A unidirectional fabric is made of continuous fibers orientated in the warp direction. For practical purposes, small weft yarns are used to group the fibers in yarns and link the yarns within a ply.

This work is part of an investigation program undertaken in the aim to supply composite manufacturers with liable data on the characteristics of the porosity within the fiber architecture. The knowledge of the space available for the resin within the preform as a function of the lay-up and the desired fiber volume fraction may greatly contribute to ameliorate the process route. A fibrous reinforcement is generally described in the producers catalogues by the characteristics of the fibers (radius, weight by unit length, number in a bundle) and the dry fabric (type and weight per unit area). But these information are of no concern for the composite manufacturers who need sound data on the compressed fibrous medium during resin infiltration. A few works were reported on the rearrangement of fibrous canvas in the stacking, the compression and the resin infiltration stages, but the results are contradictory (Gauvin *et al*., 1994; Chen *et al.*, 2001; Cadinot, 2002) and hardly applicable to materials processing. Moreover, these investigations focus on the reinforcement although the resin infiltration relates to the space between the fibers.

In the present study, we investigate the micro-structure within and around the glass fibers bundles of unidirectional carbon/polyester laminates using stereology, in order to define, without modeli-sation, more sophisticated parameters than the classical fiber volume fraction.

## MATERIAL

The composites elaborated during this work are based on a stacking of parallel plies of unidirectional glass fibers fabrics. In these fabrics the small weft yarns, which usually link the fibers, have been replaced by small stitching yarns. These particular fabrics can then be classified as non-crimp stitched fabrics (Lomov *et al.*, 2003) more than woven ones. The stitching yarns are intended to avoid the effect of waviness of the fibers encountered in classical warp knitted unidirectional fabrics.

The mean diameter of the fibers is 17.4 µm and each yarn is made of roughly 2000 fibers. A ply is characterized by its areal density (646 g/m$^2$) and thickness (0.5 mm in loose state). The glass yarns of the reinforcement are linked by chain stitched polyester yarns (3 g/m$^2$), the distance between which is approximately 11 mm. One over two consecutive polyester yarns is stitched with a small glass yarn (3 g/m$^2$) as it is schematically presented in Fig. 1.

The polymer matrix of the composite is an unsaturated polyester resin (Enydyne D 20-5361 TAS, Cray Valley) combined with a M.E.K.P. catalyst (Peroximon K1). During the polymerization lap time the impregnation of the reinforcement is made in two steps: i) the filling of the plies by the resin, ii) the compression of the whole set of superimposed wet plies.

The desired final thickness of the composite is 3 mm. The combination of the number of plies and the pressure applied during the process provides a series of composites with several glass fiber volume fractions. The experimental conditions used for a 5-plies composite lead to a fiber volume fraction of 0.441. A plane cut through this composite is represented in Fig. 2.

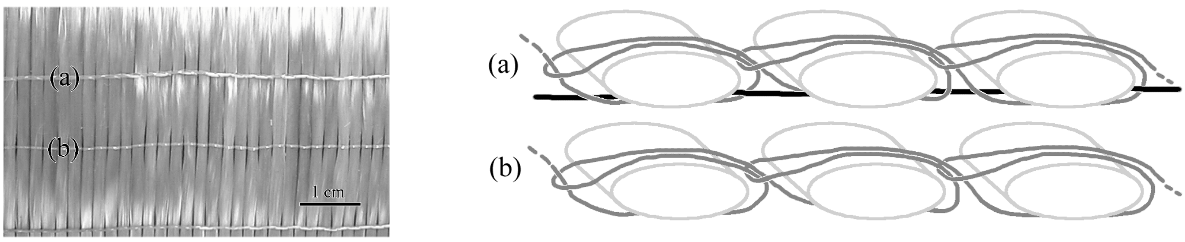
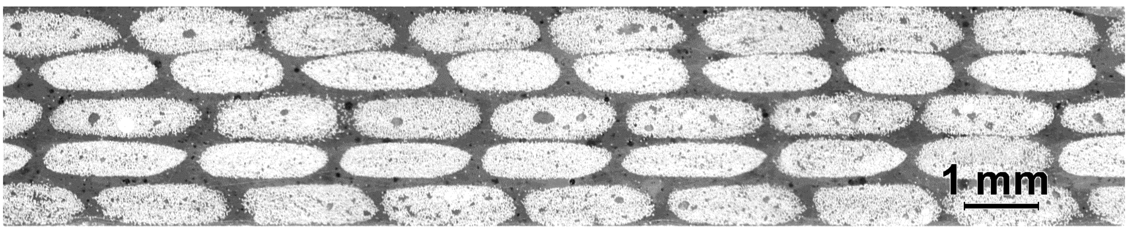
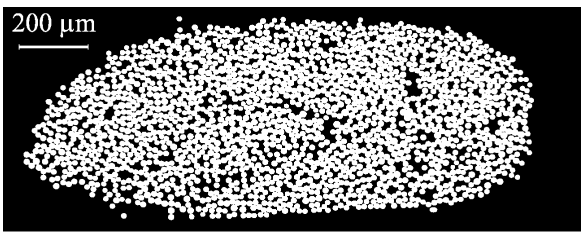



Fig. 1. *Lower face of a unidirectional ply (left) and schematic description of the two types of stitching yarns, (a) and (b), occurring alternatively along the ply.*




a)




b)

Fig. 2. *Dual-scale microstructure of an unidirectional composite containing 44.1 vol.% of glass fibers. (a) The yarns appear in light grey color and the polymeric matrix is the dark one. (b) Plane cut through a yarn.*

# EXPERIMENTAL PROCEDURE AND RESULTS

On Fig. 2a we can observe that the yarns in the upper ply are darker than those in the ply immediately below; moreover, their cross-sections look larger. The same situation is observed for the two consecutive lower plies. The darker color of some yarns suggests the existence of a higher ratio of polymeric matrix in these yarns. Several parameters can explain this heterogeneity: the location of the cutting plane in the composite, the compression applied to the yarns during the manufacturing process... From the analysis of 239 yarns sections in the material it follows that there are noticeable differences between cross-sections areas issued from separate plies but there is no systematic variation of the cross-sections areas through the thickness of the composite. This last point discards the influence of the stress applied during the process. As the number of fibers remains the same from one ply to another, the heterogeneity may be due to the presence of the stitching polyester yarns. In order to check this hypothesis it is necessary to analyze the composite along the direction of the glass yarns.

## SERIAL SECTIONS

A three dimensional exploration of the micro-structure of isolated yarns by serial sections has been tuned. The elementary volume of observation is 0.5×mm×2 mm×11 mm. The depth 11 mm corresponds to the distance between two stitching yarns. The area 0.5 mm×2 mm is the minimal rectangular area around a yarn as can be seen on Fig. 2b. For microstructural examination the sample is embedded in an epoxy resin and mounted on a plane metal base for successive grindings (Fig. 3). A thickness between 10 µm and several mm can be removed with a grinding machine. Then the sample is polished on SiC grain inlaid papers (1200, 2400 and 4000 grades) and 1µm grade diamond paste. The thickness of the removed layer, typically 1 mm, is measured by means of a tri-dimensional measurement apparatus (Tempo from MCA7). The precision on Z values is about 10 µm.

Two yarns issued from two distinct plies are chosen far from the edges of the composite and followed along the Z direction. Each plane section is observed by using a Scanning Electron Microscope (SEM) at low magnification (×70) and digitized on a square grid. The SEM is preferred to the optical microscope due to a better contrast between phases. Twelve serial sections through one yarn are presented in Fig. 4.

## GLOBAL ANALYSIS: GEOMETRY OF THE YARNS

From the images of Fig. 4, the following qualitative observations can be made: i) the extension of the yarn spans two extrema as a function of Z, ii) two different shapes are observed for the sections of minimum size (this is confirmed on the second set of sections that has not been reproduced here).

These observations are quantified with the "perimeter", P, and the "area", A, of the yarn for each section. Several definitions of these parameters can be given in the case of a yarn (from a convex envelope, from maximal Feret diameters, etc). However, they can be defined after simple morphological transformations on the digitized images of the microstructure: a closing is made on the fiber phase of the yarn by using the Aphelion software. As can be seen on Fig. 5, P decreases and A increases when the size of the structuring element increases but the variations become negligible beyond a minimum size, *i.e.*, roughly two times the fiber size. This closing size, measured from the yarn section possessing the lowest surface fraction of fibers, is then used for all yarn cross sections. After closing the yarn appears as a unique connected object containing a few holes that can be easily removed. The area of the yarn is measured on this image (*cf.* Fig. 6). Then, the difference between this set and its corresponding eroded set of size one defines a perimeter of the yarn (*cf.* Fig. 6).

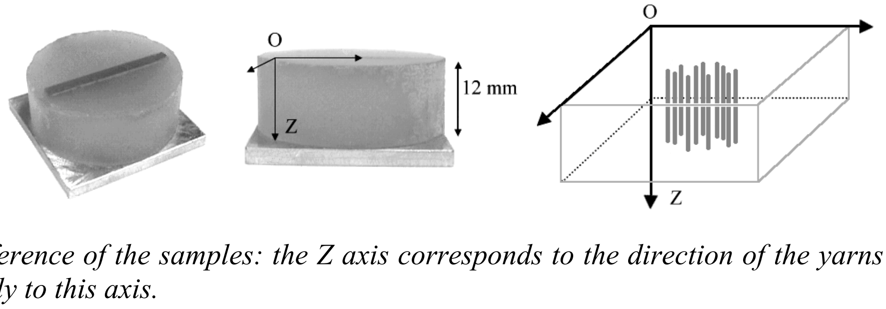



Fig. 3. *Spatial reference of the samples: the Z axis corresponds to the direction of the yarns and the sample is cut perpendicularly to this axis.*

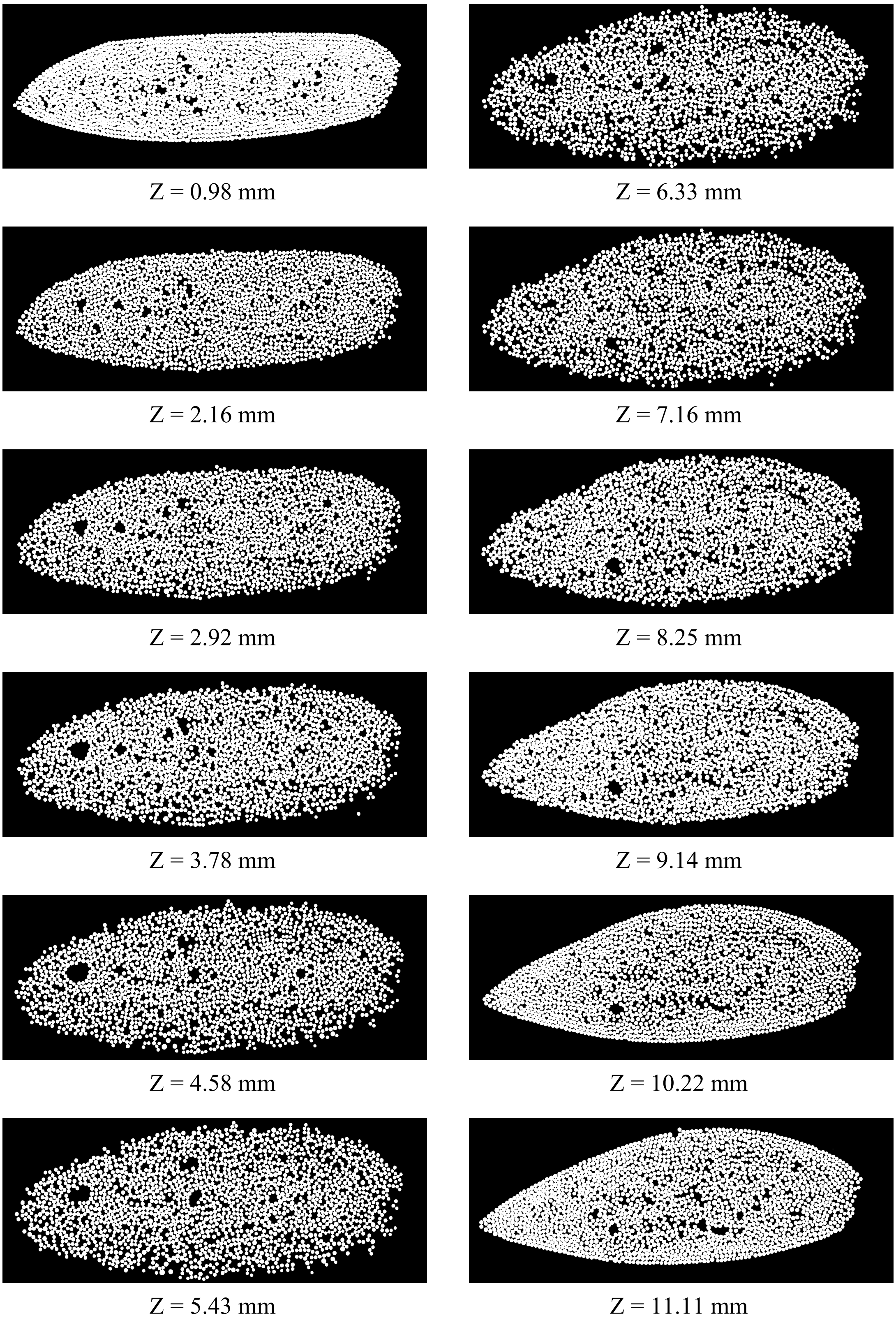



Fig. 4. *Serial sections through a yarn along the Z axis (the width of the yarn is approximately 1.5 mm).*

The results are presented in Figs. 7 and 8 for the two yarns studied. P and A follow the same evolution as a function of the altitude Z of the cutting plane. Very large variations of these parameters are observed for both yarns and the distance between the extrema of the curves is directly correlated with the distance between the polyester stitching yarns (11 mm). If the section areas are normalized by their respective mean values (0.62 mm$^2$ and 0.60 mm$^2$), the two yarns can even be described simultaneously by a unique curve presented in Fig. 9. The period of this curve, 22 mm, is two times the distance between the stitching yarns. The two minima can be attributed to the two types of stitching polyester yarns the reinforced one leading to the lowest value (*cf*. Fig. 1). A further study is necessary to check whether this evolution is restricted to the periphery of the yarn. The analysis of the yarns at higher magnification provides an answer to this question.

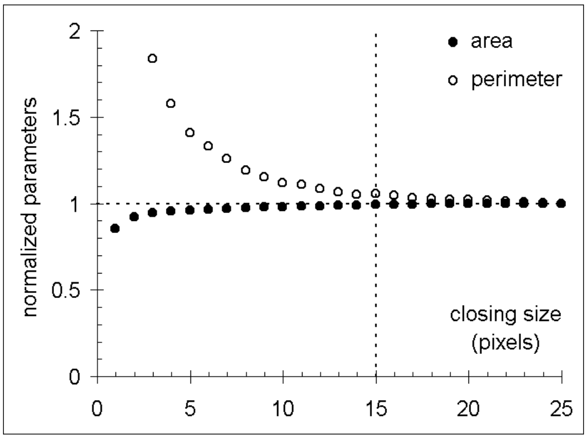



Fig. 5. *Perimeter and area of the section of a yarn as a function of the closing size (the perimeter and the area are normalized by their respective steady state values).*

Fig. 6. *Surface and perimeter of a yarn defined by morphological transformations.*

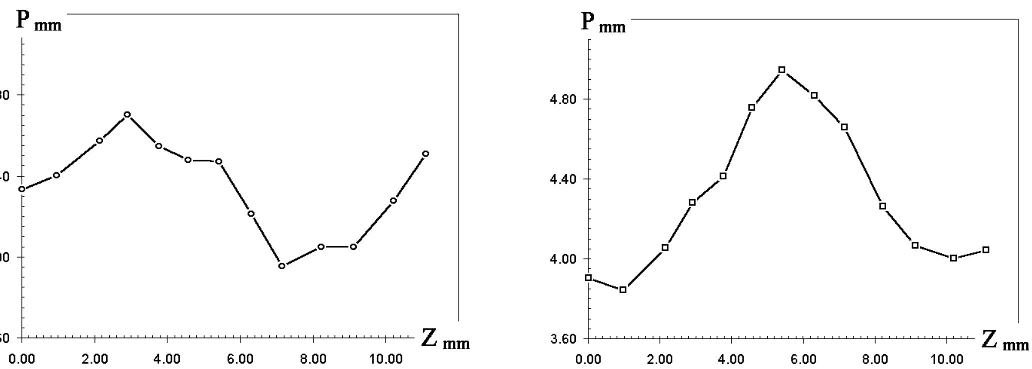
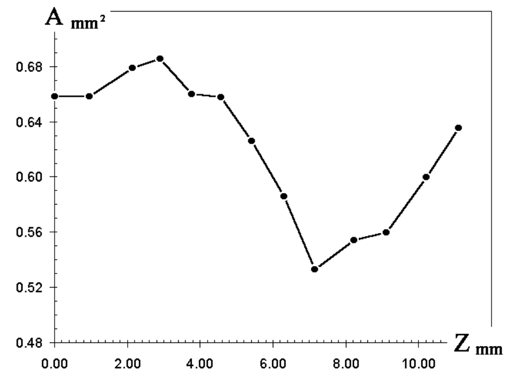



Fig. 7. *Perimeters of two different yarns as a function of the position of the cutting plane along the Z axis.*



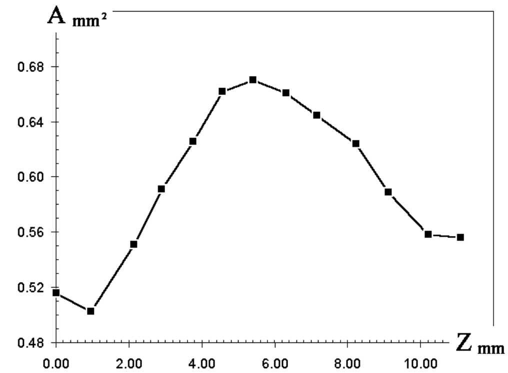




Fig. 8. *Areas of two different yarns as a function of the position of the cutting plane along the Z axis*.

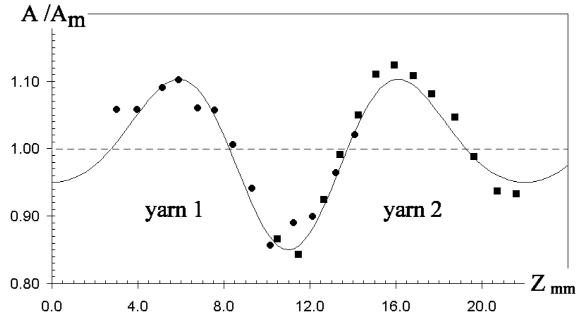



Fig. 9. *Heuristic description of the section areas through both yarns (normalized by their respective mean values).*

## LOCAL ANALYSIS: MICROSTRUCTURE OF THE YARNS

For each serial section, one field of analysis covering 30% of the yarn area is centered at the barycentre of the yarn. Let P(M,l,α) the probability for a segment of length l and direction α to be included within the matrix phase M. This probability can be calculated as a function of l from linear erosions of the matrix phase (Serra, 1982) and the following parameters can be derived:

i) the areal fraction of the matrix phase, $A_A(M)$;

ii) the mean free path, weighted in number, through the matrix phase,

$$\overline{L(M,\alpha)} = -P(M,0,\alpha)/P'(M,0,\alpha)$$

where $P'(M,0,\alpha)$ is the derivative of $P(M,0,\alpha)$;

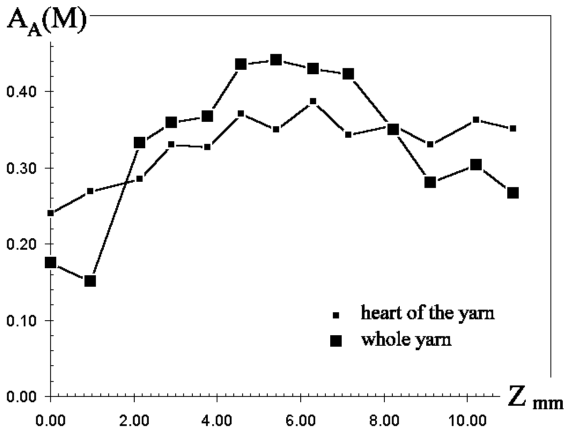
iii) the star function in 2D space,

$$St_2(M,\alpha) = [2\pi/P(M,0,\alpha)] \int_0^\infty l.P(M,l,\alpha).dl.$$

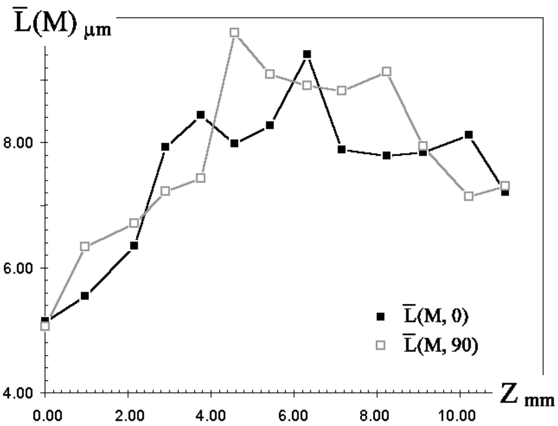
That function can be explained in the following way: from a point x of the matrix phase M let us draw a straight line through M between x and the fibrous phase; when the direction of that line describes all the space we sweep the whole area of M directly seen from x; the star function in 2D space, weighted in measure, corresponds to the mean area directly seen from all points x of M.

The linear erosions have been made along the four directions α = 0°, 45°, 90° and 135° in order to reveal a possible anisotropy. The direction α = 0° corresponds to the major axis of the section of the yarn, *i.e.*, it is contained in the plane of the ply. The direction α = 90° is parallel to the stacking direction, *i.e.*, to the direction of the compressive stress applied during the elaboration process. The measurements do not present significant differences between the four directions and only the results for α = 0° and 90°, respectively $\overline{L(M,0)}$, $\overline{L(M,90)}$, $St_2(M,90)$ and $St_2(M,90)$, have been reported here.

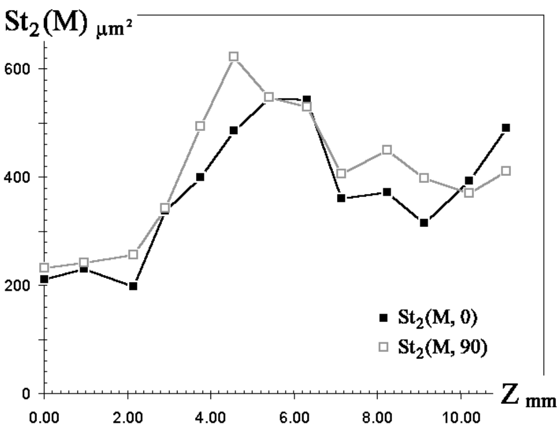
The experimental results are presented in Fig. 10. At first, the values of $A_A(M)$ calculated from this local analysis are compared in Fig. 10a with those deduced from the whole yarn (*i.e.*, not from the restricted field of analysis but from the whole yarn area). The variations of $A_A(M)$ are less pronounced for the heart of the yarn than for the whole yarn. This can also be observed in Fig. 4a. Nevertheless the evolution of the mean free path and the star function in Figs. 10b and 10c are comparable to the previous observations made for the area and the perimeter of the whole yarn.




(a)




(b)




(c)

Fig. 10. *Microstructural parameters of the polymeric phase in one yarn as a function of the position of the cutting plane along the Z axis: (a) Areal fraction (b) Mean free path (c) 2D star function.*

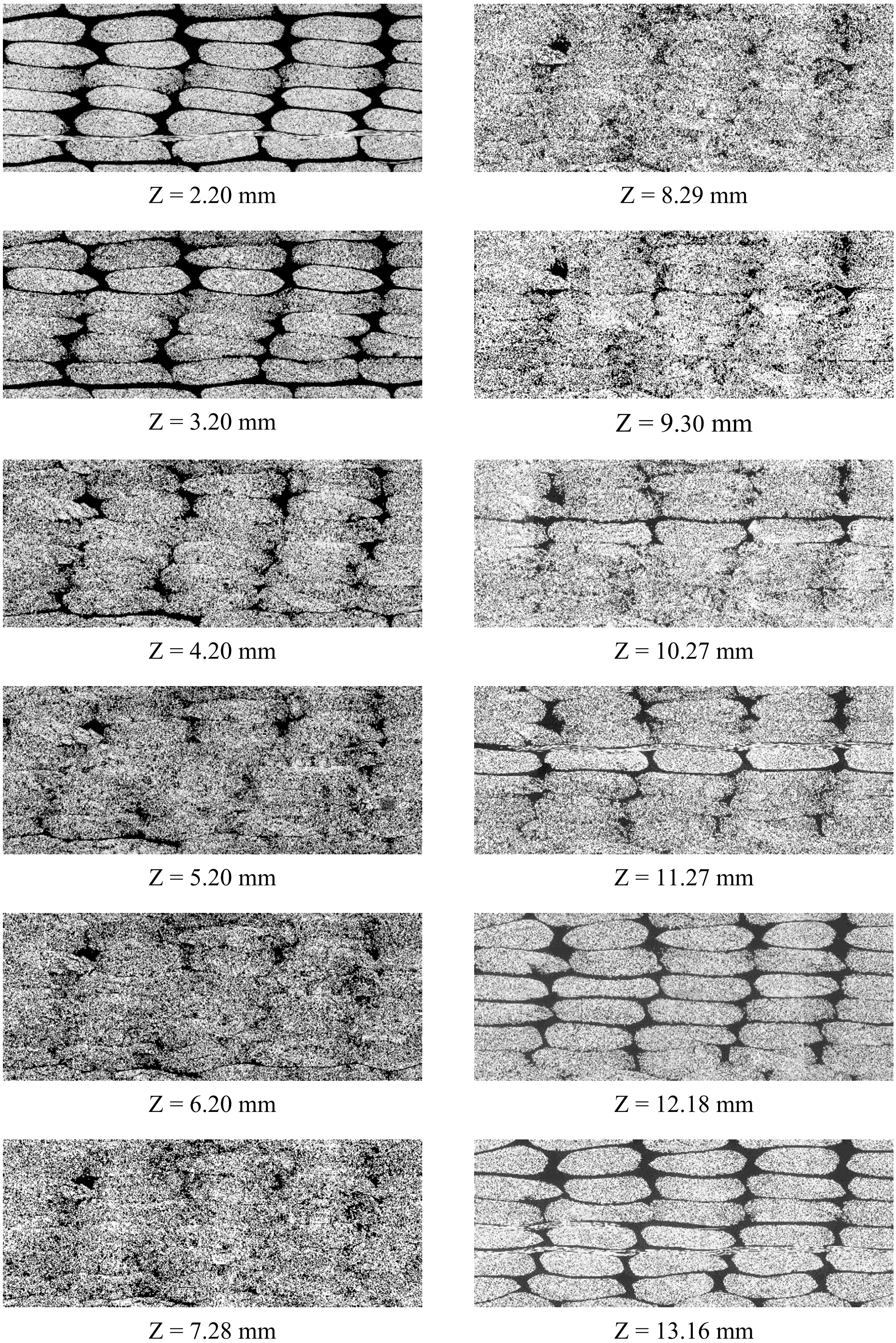



Fig. 11. *Serial sections through a R.T.M. sample along the Z axis (the volume fraction of glass fibers is equal to 0.55 and the thickness of a layer is approximately 0.4 mm).*

## SERIAL SECTIONS THROUGH A R.T.M. COMPOSITE

Finally the analysis performed on a yarn could be undertaken on the whole microstructure of a composite. In another sample elaborated by Resin Transfer Molding (R.T.M.), the stitching yarns of the successive plies have been manually superimposed. In this particular case the areal fraction of the matrix phase between the yarns (dark phase) exhibits two maxima as can be seen on Fig. 11. The small glass yarns associated with one over two stitching yarns (schematically presented in Fig. 1a) are experimentally observed on a few images in this figure. The maxima are again closely related to the distance between two stitching yarns: the area of the matrix phase between the yarns is maximum in the vicinity of the stitching yarns.

# DISCUSSION

Starting from an anodyne experimental observation (variations in the color and in the cross-sections of the yarns) serial cuts have been performed through an unidirectional composite. The microstructure of the successive sections through the yarns has been quantified using global parameters and linear morphological analysis. From the results obtained along the direction of the yarns it is obvious that the small perpendicular stitching yarns induce large variations in the fiber spacing within the yarns. These variations are conversely observed for the matrix phase between the yarns. To our knowledge, the influence of the stitching yarns on the microstructure of unidirectional composites had never been examined.

One of the main effects of the stitching yarns is the existence of a double-scale porosity. This is of first importance for the liquid molding process as the macroporosity (around the yarns) controls the permeability (Choi, 1998; Bizet *et al*., 2004). Moreover, by their influence on the yarn cross sections, the stitching yarns will modify the interactions between flows in macropores and micropores. This study affords a more precise description of the structure than does the fiber volume fraction. The macroporosity can be modelised indirectly with the perimeter and area of the yarns and the microporosity with the mean free path or the star function. Therefore, the microstructural parameters needed for the simulation of the liquid molding process are available.

Concerning the mechanical properties, the influence of the weft is well known for bi-directional woven fabrics as its overlapping with the warp induces localized residual stresses which act to depress the first damage threshold and promote further damage development. For unidirectional non crimped stitched composites the stitching yarns influence the cross section areas of the glass fiber bundles and the width of the matrix phase between them. Increasing the distance between the stitching yarns reduces the density of matrix rich domains of the laminate. That results in a higher mechanical damage threshold especially under cyclic loading conditions. Then, the inter-spacing between the stitching yarns should be as large as possible in the practical limits of the handling.

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