

# Preparation and tensile mechanical properties of unidirectional liquid crystalline single-polymer composites ☆

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## Abstract

The present study is focused on the preparation and characterization of new single-polymer composites based on liquid-crystalline fibres. Composites were obtained with two commercial wholly aromatic polyester liquid-crystalline fibres (Vectran® M and Vectran® HS) having the same chemical composition but markedly different physical properties. In particular, single-fibre tensile tests performed on Vectran® M and HS fibres evidenced moduli of 83.7 and 88.8 GPa and Weibull scale parameters of 1309 and 3374 MPa, respectively. Moreover, on the basis of DSC analysis considerably different melting signal resulted for Vectran® M and HS fibres, with peaks located at 276 and 315 °C, respectively.

Unidirectional composite preforms with 50 vol% of Vectran® M and 50 vol% of Vectran® HS fibres were produced by filament winding. In the subsequent consolidation stage the pressure and temperature conditions were properly selected in order that one component (Vectran® M) formed a continuous matrix while the other one (Vectran® HS) retained its fibrous form and most of the original mechanical properties. In this study, the effects of the consolidation pressure (in the range from 1.8 up to 8.8 MPa) and temperature (from 260 up to 285 °C) on the composite properties were investigated by density measurements, microscopic observations of the cross-sections, and mechanical tensile tests.

At a consolidation temperature of 275 °C, a minimum void content value and an optimum in the tensile properties were detected for a consolidation pressure of 4.4 MPa. At this consolidation pressure, the effect of the consolidation temperature was to increase the composites density and to improve the matrix-related tensile properties, such as the transversal modulus (from 0.5 to 1.1 GPa) and the transversal strength (from 1.3 to 3.5 MPa). On the other hand, the longitudinal tensile modulus was practically independent of the consolidation temperature in the range from 260 up to 280 °C with an average value of about 58 GPa, while it markedly decreased at 285 °C. Concurrently, the longitudinal tensile strength decreased from 920 to 480 MPa as the consolidation temperature rose.

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## 1. Introduction

Single-polymer composites are materials in which both the reinforcing (fibres) and the continuous (matrix) phases are polymers with the same chemical composition. One of the main possible advantages of this composite materials

is that strong and stable interfaces could be naturally produced since the two phases are of identical chemistry. Another important advantage of single polymer over traditional composites is the enhanced end-life recyclability that can be achieved by using the same polymer for both fibre and matrix phases. This aspect is particularly desirable to satisfy the new environmental legislation that is currently targeting high volume industries. In fact, unlike traditional heterogeneous composites (such as glass or carbon reinforced polymer composites), single-polymer composites can be entirely melted down at the end of the product life for recycling. The first example of this kind of composites

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was reported by Capiati and Porter [1], who introduced the concept of single-polymer composites by successfully preparing polyethylene/polyethylene (PE/PE) composites. Following their pioneering work, a great deal of effort has been focused on PE/PE composites prepared both with continuous [2–32] and short polyethylene fibres [33–36]. Other examples of this particular class of materials are based on polypropylene [37–43], poly(ethylene-terephthalate) [44–46], poly(ethylene-naphthalate) [47], and liquid-crystalline copolyesters [48,49] single-polymer composites. In most cases, single-polymer composites have been obtained through the so called “hot compaction” technique [50,51]. In this processing procedure, a fraction of the surface skin of an array of oriented polymer fibres or tapes is melted under relatively low contact pressure and the structure is then consolidated by the application of a substantially higher pressure for a short time. On cooling, the recrystallized polymer acts as a binder, like the resin matrix in a conventional heterogeneous composite. The use of temperature and pressure to control the melting of the outer surface layer of polymeric fibres implies that the processing window for the hot-compaction process is quite narrow. Moreover, with this approach it is very difficult, if not impossible, to preserve the original mechanical properties of the polymeric fibres. Other processing techniques reported in the scientific literature include film stacking [2,3,5,20,26,35], powder impregnation [17], and solution impregnation [15]. When both the matrix and the reinforcing phases are in a fibrous form an alternative and potentially interesting technique for the production of single-polymer composites is the co-woven and the commingled yarns techniques [52]. Both the techniques consist in the preparation of a preform either by a co-woven textile or by a commingled yarn produced by combining both phases in the same yarn. In a second step, the preform is consolidated under pressure at a temperature such that the fibrous matrix melts and flows thus forming a continuum phase that binds the reinforcing fibres. In order to be applied for the production of single-polymer composites, these techniques require the selection of suitable polymeric fibres having the same chemical nature but different melting points.

Aim of this paper is to investigate the possibility to prepare new single-polymer composites based on liquid-crystalline wholly aromatic copolyesters fibres (Vectran® [53]). Unidirectional composites have been obtained starting with commercial as-spun or thermally treated Vectran® fibres characterized by markedly different melting points. The different thermomechanical behaviour of the selected Vectran® fibres allowed us to successfully adopt a co-woven/hot consolidation process for the composites production.

## 2. Experimental

### 2.1. Materials

Composites were prepared using commercially available Vectran® fibre that is a wholly aromatic copolyesters

liquid-crystalline polymer (LCP) by Celanese Advanced Materials Inc. (now Kuraray America Inc.). Two types of fibres have been used and namely:

- Vectran® M, linear density of 750 denier (1 denier = 9 g/km), 150 filaments per yarn;
- Vectran® HS, linear density of 1500 denier, 300 filaments per yarn.

A survey of the thermomechanical properties of these fibres can be found on the article of Beers and Ramirez [53] or on the product data sheet [54]. It is worthwhile to note that these two type of fibres possess the same chemical nature, but considerably different properties: in particular melting points of 276 °C for Vectran® M and 330 °C for Vectran® HS are reported [53]. Vectran® M are as-spun fibres while Vectran® HS fibres have been subjected to a proprietary heat treatment under tension [55].

### 2.2. Composites preparation

Unidirectional composites were prepared in a two stages process, as schematized in Figs. 1 and 2. At first, both Vectran® M and HS as received fibres tows were wound on an open metal frame with an opening of about  $115 \times 175 \text{ mm}^2$ . As evidenced in Fig. 1a, this mandrel was partly collapsible in order to facilitate the subsequent consolidation stage. The winding process (see Fig. 1b) was realized by a three controlled axes filament winding machine model ALAB 0102 by T.EL.MEC. s.r.l. (Olginate LC, Italy). In order to obtain composite preforms with a 50 vol% content of both type of fibres, the winding sequence has been developed as schematized in Fig. 1c. The mandrel was then removed from the winding machine and the preform was located between two 1 mm thick PTFE sheets, and sandwiched between two  $100 \times 170 \text{ mm}^2$  aluminium plates (see Fig. 2a). The consolidation stage was performed according to the processing scheme described in Fig. 2b, where the temperature profile was experimentally determined by a thermocouple in contact with the composite. The heating stage up to the consolidation temperature  $T_c$  (in the range from 260 up to 285 °C) was performed under a constant pressure of 1.8 MPa. When the consolidation temperature was reached, the load was suddenly increased up to a consolidation pressure  $P_c$  (in the range from 4.4 to 8.8 MPa). This pressure was hold for a constant time of 30 s after which the pressure was decreased down to 1.8 MPa and the composite was allowed to cool under load. The press used for composite fabrication was a 10 tons hydraulic Carver Laboratory Press. Depending on the consolidation temperature and pressure, the resulting composite plates had thickness values in the range from 0.67 to 0.82 mm.

At a preliminary stage, some tests were also performed on Vectran® M fibres compacted under the same

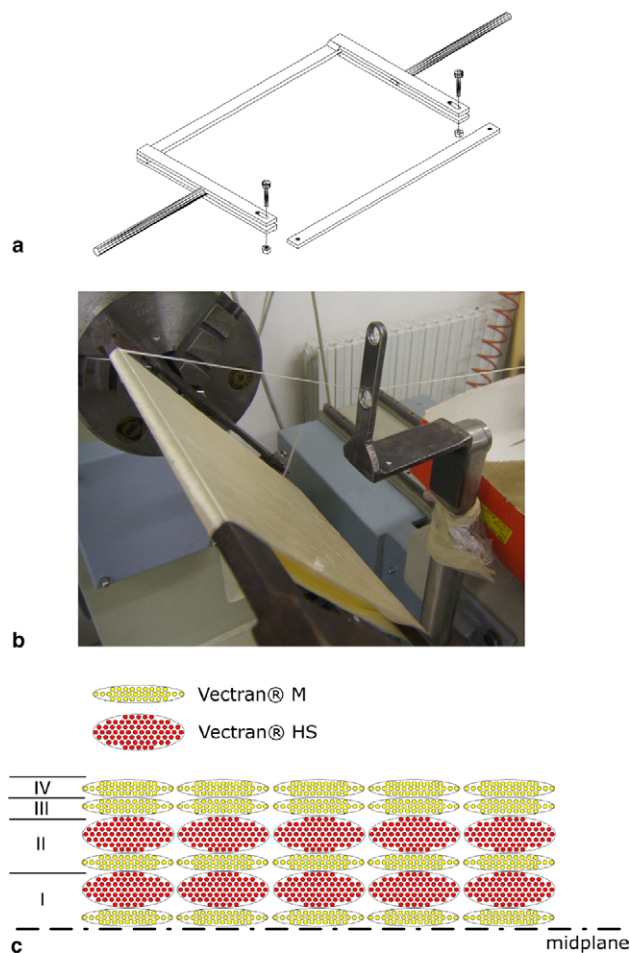


Fig. 1. Schematic of the manufacturing process of the co-woven perform by filament winding: (a) collapsible flat open square frame; (b) filament winding stage; (c) sequence of fibres deposition.

processing conditions adopted for the composites production.

### 2.3. Testing procedures

Differential scanning calorimetry (DSC) measurements were conducted by a Mettler DSC 30 calorimeter. The heating rate was 10 °C/min and all tests were conducted in nitrogen flushing at 150 ml/min.

Density measurements were performed by a Micromeritics helium pycnometer model 1330TC. The sample holder had a volume of 10 cm<sup>3</sup> and its temperature was controlled by a thermostatic bath at 23.0 ± 0.3 °C. For each sample 90 measurements were performed at a run fill pressure of 135 kPa and at an equilibration rate of 35 Pa/min.

Tensile test on single fibres and on composites in the transversal direction were performed by an Instron model 4502 universal testing machine. According to ASTM standard D3379, monofilaments were randomly extracted from a bundle and center-line mounted on win-

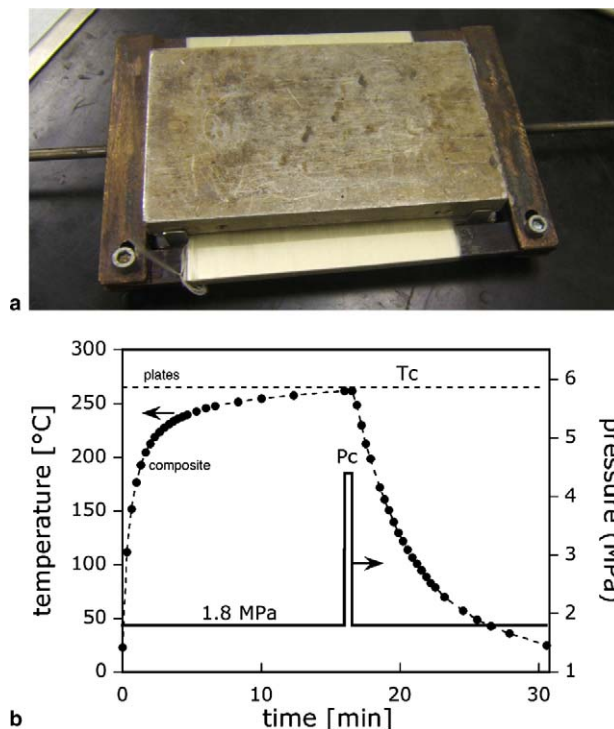


Fig. 2. Details of the hot consolidation process: (a) mould ready for the hot-compression; (b) schematic of the processing temperature and pressure profiles.  $T_c$  and  $P_c$  are the consolidation temperature and pressure, respectively.

dow cards using a quick-setting glue. Tensile strength and modulus were measured at room temperature at various gage lengths (25, 50, and 100 mm), with a 10 N load cell, at a strain rate of 0.04 min<sup>-1</sup> for each gage length. The diameter of each single fibre was determined by optical microscopy observations. Transversal tensile properties of unidirectional composites were evaluated on bars of rectangular cross-section 20 mm wide and with a gage length of 60 mm. Transversal samples consisting of at least three specimens were tested at room temperature with a load cell of 100 N, at a strain rate of 0.025 min<sup>-1</sup> and the collected data were corrected by the system compliance.

Due to the higher required loads, tensile tests of unidirectional composites in the longitudinal direction were performed by a 25 kN closed loop servo-hydraulic MTS 858 Mini Bionix testing machine. The initial axial displacement was measured by a strain gauge extensometer (MTS, model 634.31F-24; gage-length 20 mm). Longitudinal tensile properties of unidirectional composites were evaluated on bars of rectangular cross-section 15 mm wide and with a gage length of 40 mm. Steel tab ends were glued to aid gripping. Longitudinal samples consisting of at least three specimens were tested at room temperature at a strain rate of 0.025 min<sup>-1</sup>.

In all cases, the tensile modulus was determined as a secant value between two strain levels of 0.05% and 0.25%, as suggested by the ISO standard 527.

### 3. Results and discussion

#### 3.1. Fibre properties

The DSC thermograms of both Vectran® M and HS fibres are reported in Fig. 3, from which the markedly different thermal behaviour of the two fibres clearly emerges. The shape of the thermograms closely resembles those of as-spun and heat treated Vectran® fibres reported by Menczel et al. [56]. In particular, it is interesting to note that Vectran® M fibres are characterized by the presence of two broad endothermic signals the first of which has a peak at 276 °C that exactly corresponds to the melting temperature reported by the fibre producer [53]. On the other hand, for Vectran® HS fibre a single and well defined endothermic signal is found with a peak located at 315 °C. This temperature is lower than 330 °C which is the melting temperature indicated by the fibre manufacturer [53,54]. In any case, a temperature window of about 40 °C exists between the endothermic peaks of the two fibres that could be exploited for the processing of Vectran®-based single-polymer composites.

Also the mechanical properties of Vectran® M and HS fibres are markedly different, as it clearly emerges from their stress–strain curves reported in Fig. 4a for 25 mm long fibres. At very low strain levels the slope of the two curves is quite similar but, as the deformation increases, Vectran® HS fibre are able to bear much high stress and strain levels than Vectran® M. The stress–strain curve of Vectran® HS fibre show a clear slope increase as the deformation proceeds, thus indicating a cold-drawing process of the polymeric macromolecules during the tensile test. The values of Young's modulus, strength and strain at break of Vectran® M and HS fibres tested at several gage lengths are reported in Table 1. As expected, the tensile modulus increases as the gage length increases. The observed trend is the result of the system compliance that is more and more important as the gage length decreases. For both kind of fibres, a true fibre modulus can be evaluated trough the zero extrapolation

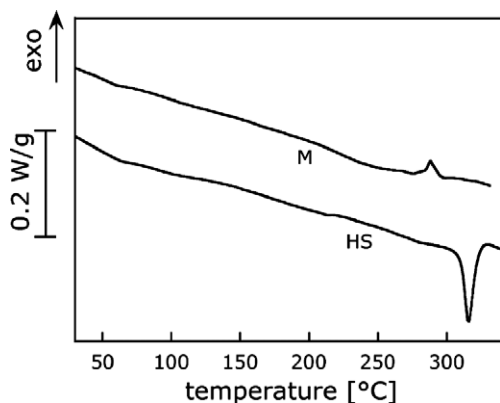


Fig. 3. DSC thermograms of Vectran® M and HS fibres.

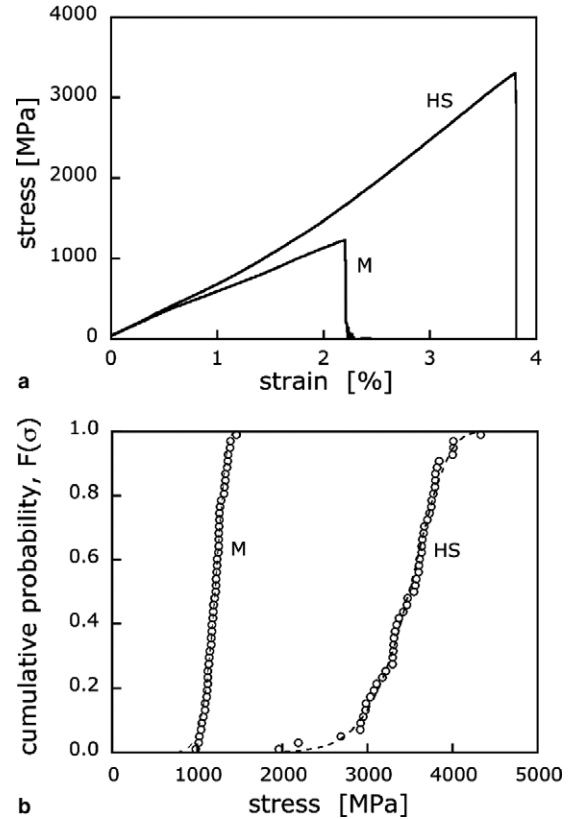


Fig. 4. (a) Tensile stress–strain curves and (b) failure probability distributions of 25 mm long Vectran® M and HS fibres.

of the inverse of the measured modulus as a function of the inverse of the gage length. The obtained values (see in Table 2) are quite similar with a slightly higher modulus for the Vectran® HS fibre. By taking into account the system compliance, the strain at break values reported in Table 2 have been also evaluated. The cumulative failure probability distributions  $F(\sigma)$  of Vectran® M and HS fibres, at a gage length of 25 mm, are reported in Fig. 4b. It clearly emerges that both fibres follow reasonably well a two-parameter Weibull strength distribution (dotted line) in the form [57,58]:

$$F(\sigma) = 1 - \exp \left[ -\frac{L}{L_0} \left( \frac{\sigma}{\sigma_0} \right)^m \right] \quad (1)$$

where  $L$  is the fibre gage length,  $L_0$  is a reference length,  $\sigma$  is the applied stress,  $\sigma_0$  and  $m$  are scale and shape parameters, respectively. It immediately appears that Vectran® HS fibre is characterized by much higher and more dispersed strength values than Vectran® M fibre. Weibull scale and shape parameters have been determined on the basis of an iterative procedure proposed by Gurvich et al. [59] in which all experimental results on specimens of different size are considered together as a statistically representative population. The obtained values are reported in Table 2. It is worth noting that Weibull scale and shape parameters allow one to estimate the average fibre strength  $\bar{\sigma}$  at a given gage length  $L$  as [58]:



Table 1  
Average values and standard deviations for the Young's modulus, strength and strain at break of Vectran® M and Vectran® HS fibres at various gage lengths

Gage length (mm)	Number of specimens	Vectran® M			Vectran® HS		
		$E$ (GPa)	$\sigma_b$ (MPa)	$\varepsilon_b$ (%)	$E$ (GPa)	$\sigma_b$ (MPa)	$\varepsilon_b$ (%)
25	49	$68.8 \pm 14.2$	$1209 \pm 108$	$2.15 \pm 0.23$	$71.9 \pm 12.7$	$3427 \pm 443$	$3.73 \pm 0.28$
50	18	$79.1 \pm 7.6$	$1149 \pm 193$	$1.73 \pm 0.26$	$79.8 \pm 9.1$	$2537 \pm 633$	$2.66 \pm 0.56$
100	18	$77.6 \pm 12.6$	$1099 \pm 242$	$1.64 \pm 0.25$	$83.7 \pm 8.1$	$2075 \pm 204$	$2.14 \pm 0.08$

Table 2  
Some properties of Vectran® fibres

Property	Vectran® M	Vectran® HS
Diameter ( $\mu\text{m}$ ) <sup>a</sup>	$23.4 \pm 1.8$	$24.0 \pm 1.4$
Density ( $\text{g}/\text{cm}^3$ ) <sup>b</sup>	$1.396 \pm 0.008$	$1.403 \pm 0.002$
Tensile modulus (GPa) <sup>c</sup>	83.7	88.8
Weibull scale parameter (MPa) <sup>d</sup>	1309	3374
Weibull shape parameter	8.28	6.13
Strain at break (%) <sup>c</sup>	1.50	1.92

<sup>a</sup> Measured by an optical microscope.

<sup>b</sup> Measured by a helium pycnometer.

<sup>c</sup> Extrapolated from the data of Table 1 by considering the system compliance.

<sup>d</sup> Referred to a 25 mm gage length.

$$\bar{\sigma} = \sigma_0 \left( \frac{L}{L_0} \right)^{-1/m} \Gamma \left( 1 - \frac{1}{m} \right) \quad (2)$$

where  $\Gamma$  is the Gamma function. It is worth noting that Vectran® HS fibre has a scale parameter, and hence an average strength, 2.6 times higher than those of Vectran® M.

Finally, as reported in Table 2, it can be observed that the two types of fibre has quite similar diameter and density values. These data are in agreement with those reported by the fibre manufacturer that indicates an average diameter of 23 microns for both fibres and a density of 1.40 and 1.41  $\text{g}/\text{cm}^3$  for Vectran® M and HS fibres, respectively.

### 3.2. Preliminary results on Vectran® M compacts

In order to optimize the preparation process of Vectran® M and HS composites, a preliminary investigation has been conducted on Vectran® M fibre compacts. Filament wound unidirectional preforms have been compacted at a consolidation pressure of 4.4 MPa at three different consolidation temperatures of 260, 275, and 285 °C. In Fig. 5a and b the tensile modulus and strength values are reported as a function of the consolidation temperature along the longitudinal and transversal directions, respectively. For as concern the longitudinal direction, when the consolidation temperature is raised from 260 to 275 °C a marked drop is observed of both modulus and strength. It is worthwhile to note that a further increase of the consolidation temperature up to 285 °C do not practically cause variations of the longitudinal tensile properties. In order to understand this behaviour it is important to remember that DSC analysis evidences a first endother-

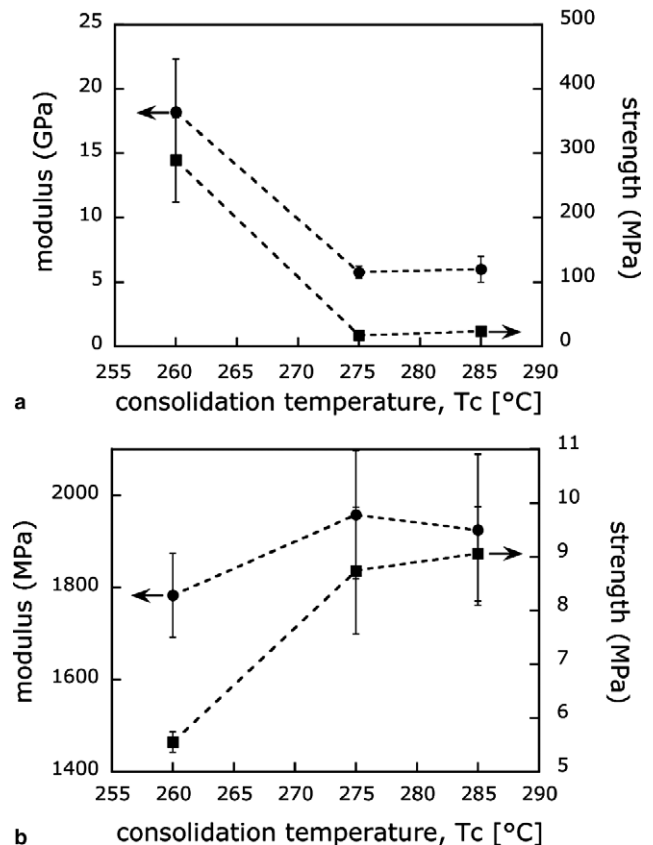


Fig. 5. Tensile modulus (●) and strength (■) of Vectran® M fibres compacted under a pressure  $P_c = 4.4$  MPa at various temperatures: (a) longitudinal direction; (b) transversal direction.

mal signal for Vectran® M fibre whose peak is located at 276 °C (see Fig. 3). This endothermal peak is quite broad and at 260 °C we already enter the melting region. In fact, the longitudinal tensile modulus at  $T_c = 260$  °C is much lower (18.2 GPa) than those of virgin Vectran® M fibres (83.7 GPa). Moreover, a consolidation temperature of 260 °C is enough to induce a transversal modulus of about 1.8 GPa. Both transversal modulus and strength improve as the consolidation temperature rises from 260 up to 275 °C. As previously observed for the longitudinal modulus and strength, also the transversal tensile properties do not substantially change when the consolidation temperature is increased from 275 °C up to 285 °C. Finally, it is worthwhile to note, that even at the highest consolidation temperature Vectran® M fibre compacts are not isotropic

thus evidencing that the material partially retains its molecular orientation.

### 3.3. Vectran® M and HS composites: effects of consolidation pressure

The influence of the consolidation pressure on the properties of unidirectional composite plates obtained with both Vectran® M and HS fibres have been investigated by keeping the consolidation temperature constant at 275 °C. A macroscopic effect of the consolidation pressure is the variation of the composites thickness. In fact, as reported in Fig. 6a, the composite thickness decreases as the consolidation pressure increases. This effect is related to the flow of the melted Vectran® M fibres toward the borders of the composite plates since they are not laterally confined in the mould. Since Vectran® HS fibre do not melt during the composites preparation we can reasonably suppose that its fibre volume fraction increases as the plate thickness decreases. The estimation of Vectran® HS fibre volume fractions is reported in Table 3. This estimation is based on the following assumptions: (i) Vectran® HS fibre volume fraction is 0.5 for the composites obtained at the lowest temperature (260 °C); (ii) the fibre volume fraction is linearly related to the plate thickness. It is important to observe that the first hypothesis has been validated by

Table 3

Effect of the processing conditions on the fibre (Vectran® HS) fraction and void content

Consolidation temperature $T_c$ (°C)	Consolidation pressure $P_c$ (MPa)	Fibre volume fraction (vol%)	Void content (vol%)
275	1.8	$51 \pm 3$	1.85
275	4.4	$53 \pm 3$	0.55
275	6.2	$53 \pm 3$	1.72
275	8.8	$55 \pm 5$	2.24
260	4.4	$50 \pm 2$	1.34
265	4.4	$51 \pm 2$	0.85
270	4.4	$53 \pm 3$	0.61
280	4.4	$57 \pm 3$	0.55
285	4.4	$62 \pm 6$	0.75

direct microscopic observation of the composite cross-section (see Section 3.4). As reported in Fig. 6b, the density of the composites shows a non-monotonic trend with the consolidation pressure. In fact, quite unexpectedly, the density of the composites reach a maximum value for 4.4 MPa. This behaviour is not completely clear yet. A possible explanation relies on the hypothesis that at 275 °C the melted Vectran® M fibre should have a very elevated viscosity and, consequently, when the consolidation pressure is higher than a given value the flow of the matrix may cause the displacement of the reinforcing Vectran® HS fibres and the formations of voids in the composite. According to the ASTM standard D2734 the void content,  $V_v$ , of the composites can be estimated by the following equation:

$$V_v = \frac{\rho_{th} - \rho_{exp}}{\rho_{th}} \quad (3)$$

where  $\rho_{th}$  and  $\rho_{exp}$  are the theoretical and the experimental values of the composite density, respectively. The theoretical density of the composites can be estimated by using the following expression [60]:

$$\rho_{th} = \rho_f V_f + (1 - V_f) \rho_m \quad (4)$$

where  $\rho_f$  and  $\rho_m$  are the densities of fibre (Vectran® HS) and matrix (Vectran® M), respectively, and  $V_f$  is the fibre volume fraction. The obtained void content values are reported in Table 3. The void content can be taken as a parameter for the estimation of the quality of composites. In general, a good composite should have less than 1% void, whereas a poorly made one can have up to 5% void content [60]. At the consolidation temperature of 275 °C we can therefore conclude that a consolidation pressure of 4.4 MPa is suitable to produce an acceptable low void content.

At this point, it is interesting to analyse how the consolidation pressure affects the tensile mechanical properties along the longitudinal and the transversal directions. As it clearly emerges from Fig. 7a, the longitudinal tensile modulus is practically independent of the consolidation pressure and it lies around an average value of 56 MPa. The tensile modulus of virgin Vectran® HS fibre is

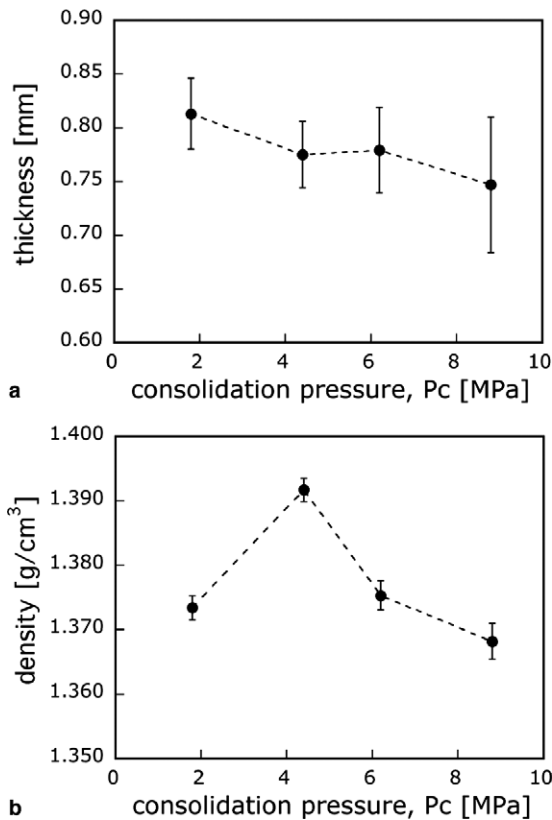


Fig. 6. Effect of consolidation pressure on: (a) the thickness and (b) the density of composite plates obtained with Vectran® M and HS fibres consolidated at a temperature  $T_c = 275$  °C.

88.8 MPa and its average volume fraction is 0.53. By a simple rule of mixture, it can be deduced that the longitudinal tensile modulus of the Vectran® M based matrix is about 19 GPa. This value is much higher than the value measured on the Vectran® M fibre compacts obtained under the same processing conditions (see Fig. 5a), thus indicating that the presence of Vectran® HS fibres influences the thermo-mechanical behaviour of Vectran® M fibres. This influence could be related to their constrain during the consolidation stage. Also the longitudinal tensile strength of the composites is not strongly influenced by the consolidation pressure, even if a slightly tendency to decrease can be noticed. For as concern the transversal direction, the dependence of the tensile properties on the consolidation pressure is summarized in Fig. 7b. It is interesting to observe that a markedly improvement of both tensile modulus and strength values occurs as the consolidation pressure increases from 1.8 to 4.4 MPa. A further rise of the consolidation pressure does not cause an appreciable variation of the transversal modulus. On the other hand, a concurrent slight decrease of the transversal strength can be noticed. Therefore, at the consolidation temperature of 275 °C it emerges that an optimal value of the consolidation pressure is around 4.4 MPa.

### 3.4. Vectran® M and HS composites: effects of consolidation temperature

As reported in Fig. 8a, the thickness of the composite plates decreases as the consolidation temperature increases. This effect can be reasonably explained by taking into account that as the consolidation temperature grows the viscosity of the melted Vectran® M fibres decreases and more matrix-material is squeezed out of the composite plates. As a consequence, the estimated values of Vectran® HS fibre volume fraction increase with the consolidation temperature as summarized in Table 3. Concurrently, as reported in Fig. 8b the composite density changes with the consolidation temperature, increasing from 1.380 g/cm<sup>3</sup> at 260 °C up to 1.392 g/cm<sup>3</sup> at 280 °C. A further temperature rise causes a slight decrease of the composite density.

Some optical micrographs of polished transverse cross-sections of composites are collected in Fig. 9. These pictures indicate that Vectran® M fibres actually melt thus forming a partially continuous phase, while Vectran® HS fibres partially preserve their original shape even if at the highest consolidation temperatures they tend to compact. Further, it is worthwhile to note that the number of voids seems to decrease as the consolidation temperature increases, accordingly to the density data.

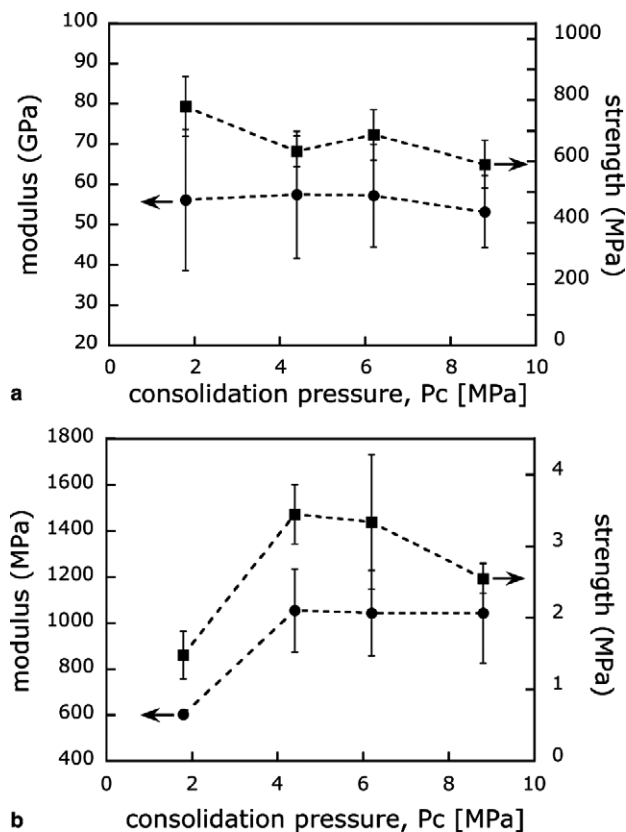


Fig. 7. Effect of consolidation pressure on the tensile modulus (●) and strength (■) of composites based on Vectran® M and HS fibres consolidated at a temperature  $T_c = 275$  °C: (a) longitudinal direction; (b) transversal direction.

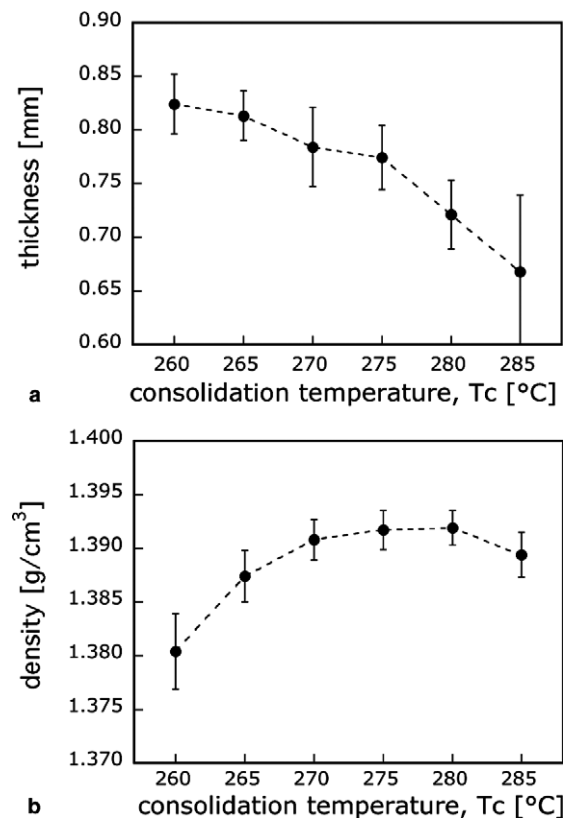


Fig. 8. Effect of the consolidation temperature on (a) the thickness and (b) the density of composites based on Vectran® M and HS fibres obtained at a pressure of 4.4 MPa.

The analysis by the line method [61] of several micrographs as those reported in Fig. 9a, indicates that Vectran® HS fibre volume fraction in the composites obtained at a consolidation pressure of 4.4 MPa and at the lowest consolidation temperature of 260 °C is  $0.50 \pm 0.02$ . This means that under this processing conditions Vectran® M matrix is not squeezed out of the composite.

The consolidation temperature also profoundly affects the tensile mechanical behaviour of the composites, both along the longitudinal and the transversal directions. In particular, it can be observed that the stress–strain curves in the longitudinal direction (Fig. 10a) of composites obtained at consolidation temperatures of 260, 265 and 270 °C show a first load drop that does not cause the complete failure of the specimens. In fact, this first peak is most probably related to the failure of one of the four Vectran® HS fibre layer in the composite. In fact, three further consecutive and very pronounced load peaks can be observed that could correspond to the failure of the remaining three plies. This behaviour could be tentatively related to the excessively low interlaminar shear strength values reached at the lowest consolidation temperatures. In fact, as reported in the second part of this paper [62], the interlam-

inar shear strength values markedly increase with the consolidation temperature. When a consolidation temperature of 275 °C or higher is used, a single load drop is observed corresponding to the complete failure of the composite. At the same time, the transversal stress–strain curves, reported in Fig. 10b, clearly evidence an increase of both elastic and ultimate properties as the consolidation temperature increases.

The tensile modulus and strength values in the longitudinal direction are reported in Fig. 11a as a function of the consolidation temperature. It can be noticed that the longitudinal tensile modulus is practically independent of the consolidation temperature up to 280 °C and that a substantial decrease can be observed at 285 °C. At the same time, the longitudinal tensile strength decreases from 920 to 480 MPa when the consolidation temperature increases from 265 to 285 °C. This trend is the results of the combined effects of: (i) the temperature dependence of the Vectran® M ed HS mechanical properties; and (ii) the increase of the Vectran® HS volume fraction with the consolidation temperature (see Table 3).

The effect of the consolidation temperature on the transversal tensile properties is summarized in Fig. 11b. As expected, both tensile modulus and strength display a tendency to increase as the consolidation temperature increases. In particular, coherently with the failure modes,

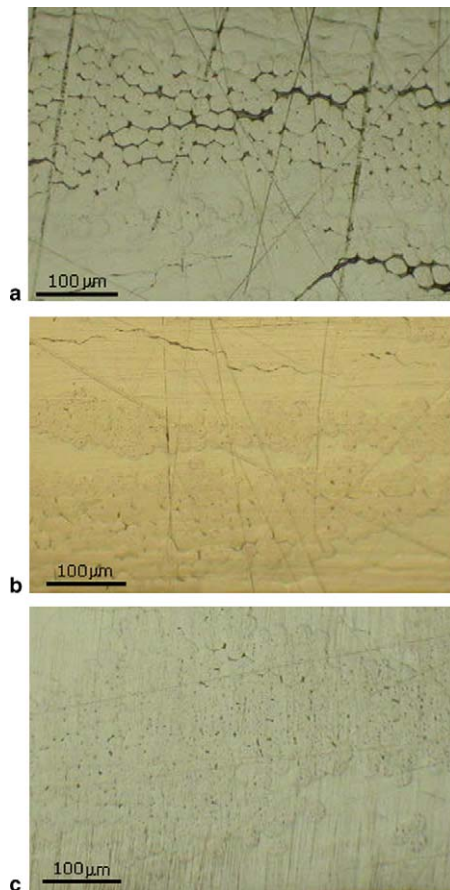


Fig. 9. Optical micrographs of polished and un-etched transverse cross sections of composites obtained with Vectran® M and HS fibres at a consolidation pressure of 4.4 MPa and at consolidation temperatures of: (a) 260 °C; (b) 270 °C; (c) 280 °C.

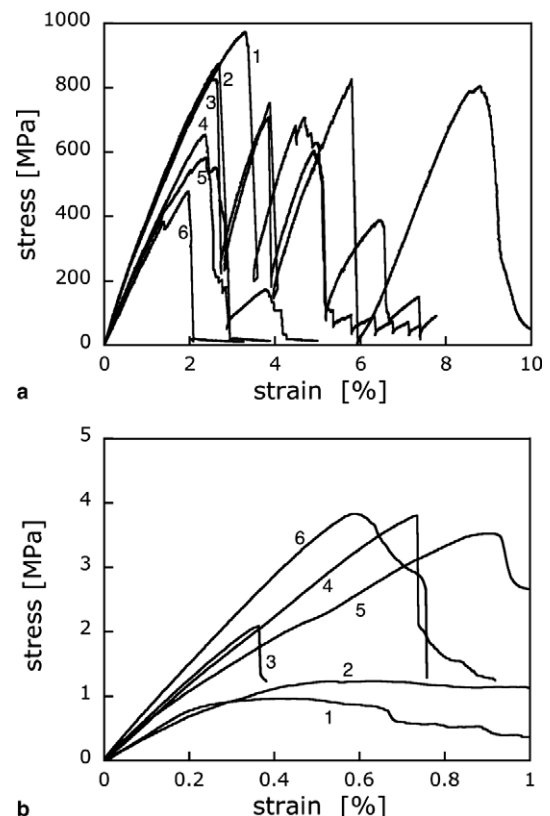


Fig. 10. Stress–strain curves along: (a) longitudinal and (b) transversal direction of composites based on Vectran® M and HS fibres consolidated under a pressure of 4.4 MPa at various temperatures: (1) 260 °C; (2) 265 °C; (3) 270 °C; (4) 275 °C; (5) 280 °C; (6) 285 °C.



it is interesting to note that the transversal strength markedly increases up to 275 °C and then it remains practically constant when the consolidation temperature is further increased up to 285 °C.

#### 4. Conclusions

A double step process has been developed for the production of single-polymer liquid-crystalline composites based on Vectran® fibres. In a first stage a unidirectional preform is realized by co-winding two different types of Vectran® fibres that possess markedly different melting points. In the subsequent consolidation stage the pressure and temperature conditions are such that one component (Vectran® M) is melting and forming a continuous matrix while the other one (Vectran® HS) retains most of its original mechanical properties.

The longitudinal tensile mechanical properties are relatively unaffected by the consolidation pressure in the range from 1.8 to 8.8 MPa, while in the transversal direction a consolidation pressure of 4.4 MPa has been proven to yield optimal values of both tensile modulus and strength.

The consolidation temperature is the key variable in the production of single-polymer liquid-crystalline composites.

In fact, as the consolidation temperature increases from 260 °C up to 285 °C, a marked improvement is observed in the matrix-related tensile properties, such as the transversal modulus (from 0.5 to 1.1 GPa) and the transversal strength (from 1.3 to 3.5 MPa). The longitudinal tensile modulus is practically independent of the consolidation temperature in the range from 260 up to 280 °C with an average value of about 58 GPa, while it markedly decreases at 285 °C. Concurrently, the tensile strength decreases from 920 to 480 MPa as the consolidation temperature rises.

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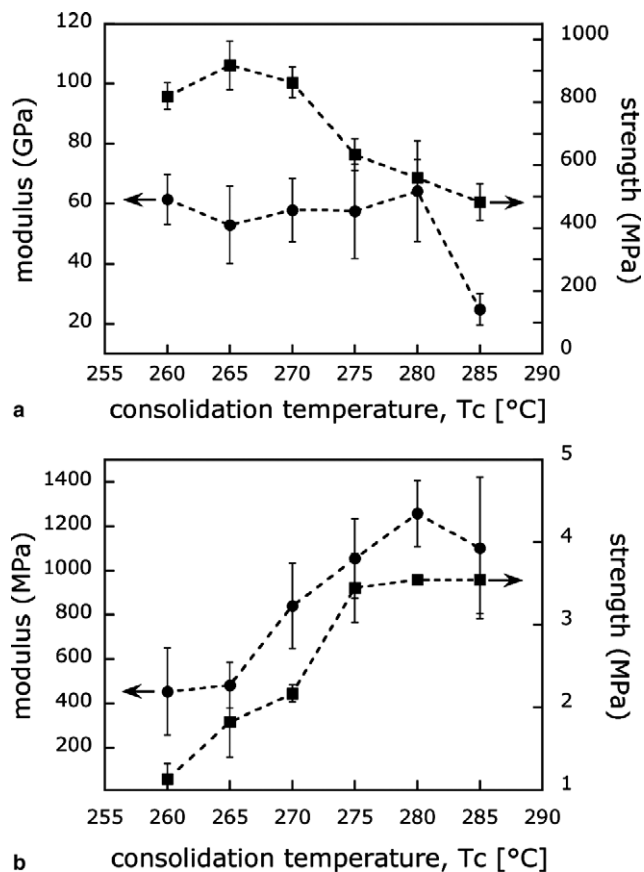


Fig. 11. Effect of consolidation temperature on the tensile modulus (●) and strength (■) of composites based on Vectran® M and HS fibres consolidated under a pressure of 4.4 MPa: (a) longitudinal direction; (b) transversal direction.

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