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Mechanical properties of Zylon/epoxy composite

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Abstract

The Zylon/epoxy composite is formed by wet-winding Zylon fibre, Poly(p-phenylene-2,6-benzobisoxazole) or PBO, with the epoxy (Stycast 1266). The effects of pre-stress on the distribution and the filling factor of the fibre are studied. It is shown that a Zylon/epoxy composite with a uniform fibre distribution and a very high filling factor is achievable. The mechanical properties of the Zylon/epoxy composite at room temperature and 77 K are investigated by uni-axial tensile tests and transverse compression tests. The results indicate that the ultimate tensile strength (UTS) of the Zylon/epoxy composite is mainly determined by the fraction of the Zylon fibre. The UTS of the Zylon fibres in the composite is found to be larger than 4.3 GPa. Due to the easy processing and the very high UTS, the Zylon/epoxy composite is suitable as reinforcement material for high-field magnet coils.

Keyword: A. Fibres; A. Polymer-matrix composites; B. Strength; B. Stress/strain curves; E. Filament winding;

1. Introduction

Non-destructive wire-wound coils for producing pulsed magnetic fields in the 80 T range require very high strength of the construction materials. The copper-based conductors available today can hardly fulfil the required mechanical properties for the coil. Therefore, internal and external reinforcements with stronger materials are necessary. These reinforcements can be realised by solid cylinders or by fibre-wound composites. The advantage of fibre-wound composites is that they can be applied easily in different thickness and in any area of the coils.

Zylon, Poly(p-phenylene-2,6-benzobisoxazole) or PBO fibre [1, 2], is a newly developed, commercially available polymer material with extremely high ultimate tensile strength (UTS), high elastic modulus and good electrical insulation. Therefore, Zylon fibre is chosen as one of the candidates for internal reinforcement of high-field magnet coils.

The main mechanical properties at room temperature of the Zylon fibre, taken from the web site of its producer (http://www.toyobo.co.jp/e/seihin/kc/pbo), Toyobo Co. in Osaka, Japan, are listed in Table 1.

Zylon fibre will be wound and impregnated with epoxy when used as reinforcement material in magnet coils. However, the information on the formation of Zylon/epoxy composite and its mechanical properties at room temperature as well as at liquid nitrogen temperature is not available. Therefore, a series of experiments, including the fabrication process of Zylon/epoxy composite, uni-axial tensile tests, transverse compressive tests and explo-vessel tests [3], have been carried out in this study. The main results related to the pre-stress effects on the distribution and the filling factor of Zylon fibres in Zylon/epoxy composite, the uni-axial tests and transverse compressive tests are presented in this paper.

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Table 1 Mechanical properties of Zylon fibre

Fibre type	Zylon HM (111 tex)*
Density [g/cm^3]	1.56
Ultimate tensile strength [GPa]	5.8
E-modulus [GPa]	280
Elongation at break [%]	2.5
Thermal expansion coeff. [1/K]	-6×10^{-6}
Dielectric constant	2.1

* 1 tex = 1gram/km

2. Experimental

The Zylon/epoxy composite sample is prepared by wet-winding the Zylon fibre with epoxy (Stycast 1266) onto a sample holder. A pre-tensile-stress is applied to the Zylon fibre bundle during winding. The sample is cured at room temperature for more than 16 hours before testing.

The sample holder for uni-axial tensile tests is shown in Fig. 1. The cross section of the racetrack-shaped sample is about 1 mm thick and 3 mm wide on both sides. A 2-element 0°/90° stacked rosette strain gage is directly attached to the side of the sample before the epoxy is hardened.

For uni-axial transverse compressive tests, two kinds of samples are prepared (Fig. 2). One is cylindrical, in which the fibre direction is in tangential direction and the applied compressive force is along the cylinder axis. The inner and outer diameters of the sample are about 8 mm and 15 mm, respectively and the height is 8 mm. The other is bar-shaped and the compressive stress is applied transverse to the fibre direction (direction 3 in Fig.2). The sample size is about 8×8×14 mm^3. Strain gages are attached to the outer surface of the sample and oriented in desired directions (cylinder axis or direction 1, 2 and 3 of the bar).

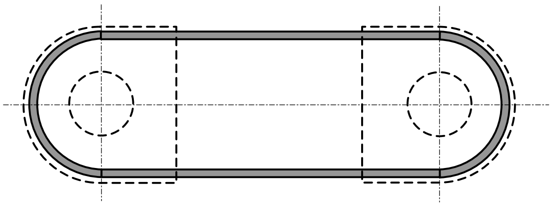


Fig.1 The racetrack-shaped Zylon/epoxy composite sample with the sample holder (dashed line) for uni-axial tensile test.

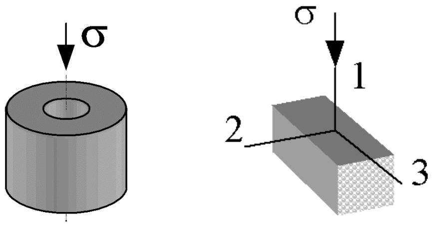


Fig. 2 Two kinds of Zylon/epoxy composite samples for uni-axial transverse compressive tests. Left: cylindrical; Right: bar-shaped.

The stress-strain measurements are carried out in a tensile test set-up. The uni-axial load is measured using a force transducer (type U9B from HBM). The experiments are conducted either at room temperature or at 77 K.

The filling factor of the Zylon fiber in a Zylon/epoxy composite sample is calculated using the formula nA_1/A, where n is the number of Zylon fibre bundles, A_1 the cross-section area of one bundle, and A the total cross-section area of the sample. The cross-section area of the bundle, A_1, is obtained using the weight of unit length of the Zylon fibre bundle (111 tex, 664 filaments) divided by the density of the fibre. $A_1 = 0.07115$ mm^2 is used for all the samples in this report.

3. Results and discussion

3.1 The influence of pre-tensile-stress during winding on the filling factor and the distribution of Zylon fibre in epoxy

Fig. 3 shows the cross sections of four Zylon/epoxy composite samples prepared with different pre-tensile stress during winding. It is found that increasing pre-tensile stress makes Zylon fibres more uniformly distributed in the epoxy matrix. The big gaps between fibre-bundles are eliminated completely under high pre-tensile stress. The filling factor of Zylon fibre increases from about 60% to more than 90% with the increase of the pre-tensile stress. Although the maximum possible filling factor for rigid rods is 90.7%, it is observed from high-magnification pictures that the cross-section of Zylon fibres can be deformed to a hexagonal shape when pushing one against another, therefore, an even higher filling factor is possible. A high filling factor is more easily obtained in the cylindrical sample than in plate-shaped one.

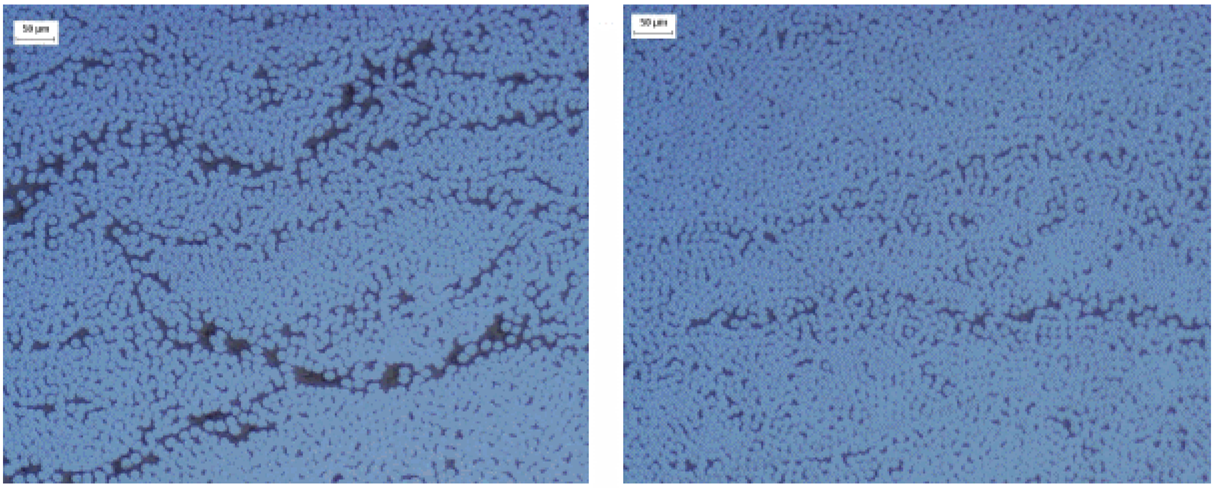
3.2 The stress-strain curve of Zylon/epoxy composite in a uni-axial tensile test

Fig. 4 shows the typical stress-strain curves of the Zylon/epoxy composite along the loading direction 3 (fibre direction) and along the transverse direction 1 under uni-axial tensile stress. The strains $-\varepsilon_1$ and ε_3 increase almost linearly with the stress before the fibres begin to break. Under uni-axial tensile stress, the material experiences mainly elastic deformation before breaking. The maximum strain along the fibre direction is about 1.7%. The ultimate tensile strength (UTS), elastic modulus and the Poisson’s ratio of this sample (filling factor = 67.3%) are 2.9 GPa, 170 GPa and 0.35, respectively.

The stress-strain curve of pure epoxy (Stycast 1266) under uni-axial tensile load is shown in Fig. 5. The UTS of the pure epoxy sample is found to be 52 MPa, about 100 times smaller than that of Zylon fibres (Table 1). Therefore, the uni-axial tensile load applied to a Zylon/epoxy composite with a high filling factor is carried almost only by the Zylon fibres. The maximal load a Zylon/epoxy composite sample can carry is found to be proportional to the number of Zylon bundles in the samples, as shown in Fig. 6. The influence of the fibre filling-factor on the maximum load is not significant. The slope of the line in Fig. 6 is about 0.31 kN per bundle, corresponding to the UTS of the Zylon fibre of 4.3 GPa. This UTS value is lower than that given in Table 1. The reasons may be the following:

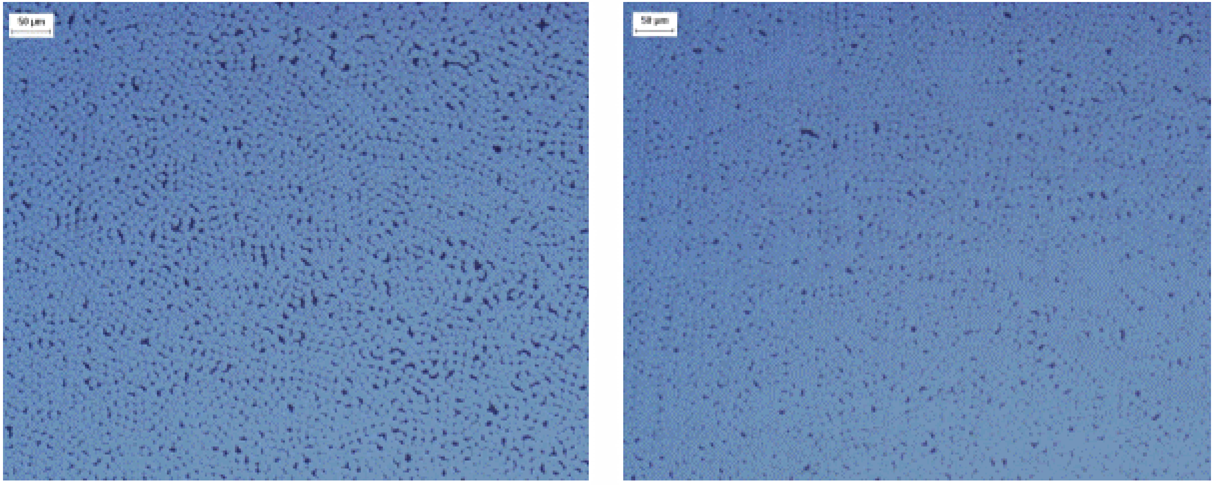
First, the tensile load is not shared uniformly by all Zylon fibres in the Zylon/epoxy composites, as in the general case of composite materials, while the UTS in table 1 is obtained using a single Zylon fibre. Secondly, the constant pre-tensile stress applied to the Zylon bundle during winding in preparing the racetrack-shaped sample results in a non-uniformly distributed residual stress, i.e. tensile stress in the outer layers and compressive stress in the inner layers after the mold is removed. The outer layer of the racetrack-shaped sample experiences a larger tensile stress (applied stress plus residual stress) in the tensile test than other Zylon layers. This is illuminated schematically in Fig. 7. Therefore, the outer layer will first reach the UTS and break, resulting in the failure of the sample. Based on this consideration, the real UTS of the Zylon/epoxy sample should be the measured UTS plus the residual tensile stress in the outer layer, which should be a fraction of the pre-tensile stress of 0.7 GPa applied to the Zylon bundle during winding.

The failure of the Zylon/epoxy composite starts locally. However, unlike the carbon- or glass-fibre composites which show a clear cut through whole cross-section at the breaking position, the fracture points of different Zylon fibres in Zylon/epoxy composite may spread over several centimetres when the composite fails. Also Zylon fibres are more flexible and can be knotted without breaking.



a). Pre-tensilestress < 70 MPa

b). Pre-tensilestress 280 MPa



c). Pre-tensilestress 700 MPa

d). Pre-tensilestress 1000 MPa

Fig. 3 Cross-sections of Zylon/epoxy composite samples, showing the effects of pre-tensile stress on the distribution and filling factor of Zylon fibre in an epoxy matrix. The size-bar is 50 µm.

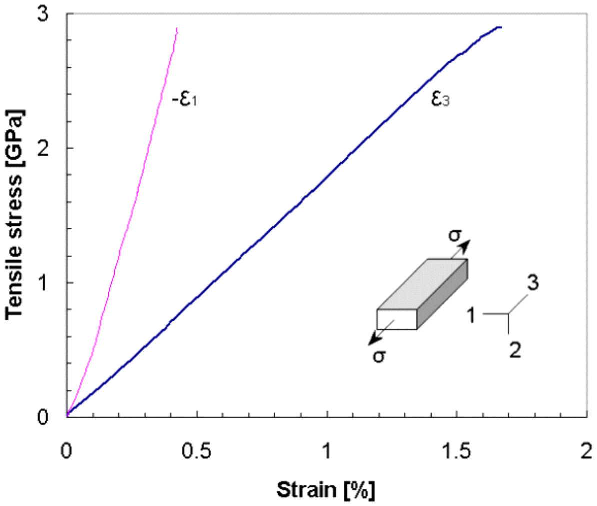


Fig. 4 Typical stress-strain curve of a Zylon/epoxy composite sample (filling factor = 67.3%).

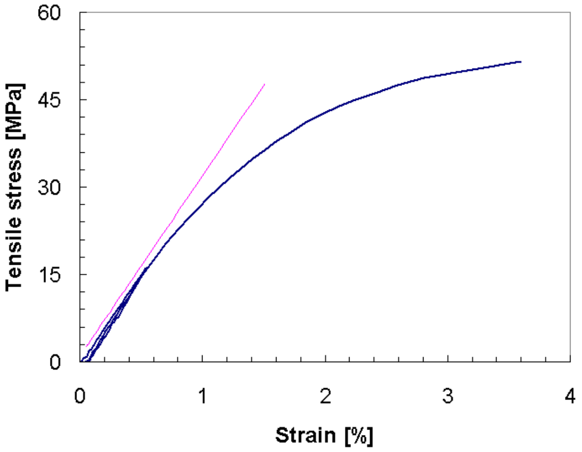


Fig. 5 Tensile stress-strain curve of pure epoxy (Stycast 1266) at room temperature. The ultimate tensile strength (UTS) and the elastic modulus are 51.6 MPa and 3.1 GPa, respectively.

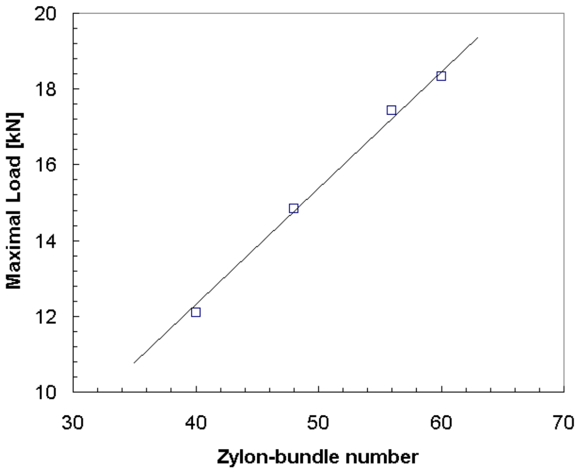


Fig. 6 The maximal load in uni-axial tensile test of the Zylon/epoxy composite samples as a function of the number of Zylon bundles in the samples (filling factor = 67.3%).

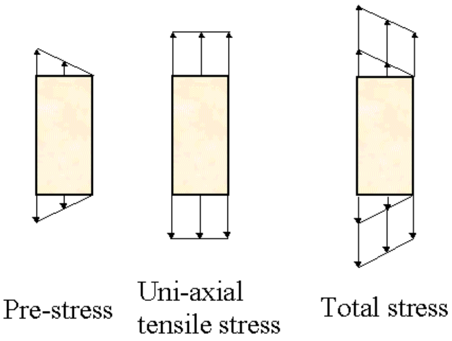


Fig. 7 The effect of pre-tensile stress applied to the Zylon bundle during winding on the stress distribution in the Zylon/epoxy sample. The left side of the rectangular box in the figure, corresponding to the outer layer of the racetrack-shaped sample, experiences a larger tensile stress in the tensile test.

At 77 K, the ultimate tensile strength, E-modulus and maximum elongation of the composite are larger than those at room temperature (Fig. 8). This is beneficial for a reinforcement material for magnet coils that are operated at liquid nitrogen temperature. The coefficient of thermal expansion of Zylon is -6×10^{-6}/K (Table 1). Therefore, the Zylon fibre will expand during cooling. The expansion of the Zylon fibres counteracts the contraction of the epoxy matrix. The result leads to the release of the residual tensile stress in the fibres. This may account for a small part of the increase of the UTS and maximal strain at 77 K in the racetrack samples used for the tensile test.

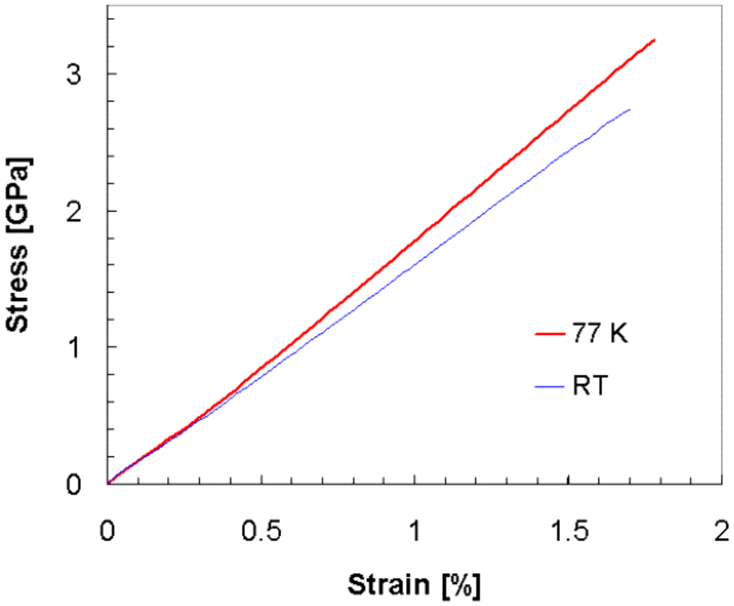


Fig. 8 The stress-strain curves of a Zylon/epoxy composite sample (filling factor = 67.3%) at two different temperatures. The test was first carried out at 77 K with the load up to 20 kN (the maximal permissible load for the test set-up) without breaking. After unloading and warming up, the sample was used for the second test at room temperature until breaking.

3.3 The stress-strain curve of Zylon/epoxy composite in uni-axial transverse compressive test

Figures 9 and 10 show the transverse compressive stress-strain curves of the Zylon/epoxy composite samples. From the results of a bar-shaped sample (Fig. 9) we can obtain the transverse compressive E-modulus E_1 as well as the other two Poisson's ratios ν_{12} and ν_{13}. E_1 and ν_{12} are found to be 2.7 GPa and 0.6, respectively. ν_{13} is very small. According to the condition of symmetry of compliances [5], we have

$$n_{13} = \frac{n_{31}}{E_3} E_1 , \qquad (1)$$

where E_3 and ν_{31} are the elastic modulus along 3 direction (the fibre direction) and the Poisson's ratio for transverse strain in the 1 direction when stressed in 3 direction. If the tensile test results of E_3 and ν_{31} are used, i.e. E_3 = 205 GPa (filling factor of 77.5%) and ν_{31} = 0.35, and E_1 =2.7 GPa from the transverse compressive test (Fig. 9), then the ν_{13} will be 0.0046 only. The Poisson's ratios $\nu_{12,}$ ν_{13} and ν_{31} satisfy the restrictions on elastic constants of orthotropic materials [5].

The slope of the stress-strain curve of a cylindrical sample is found to be about 4 GPa (Fig. 10). Unlike in the bar-shaped case, the applied axial force to an anisotropic cylinder will not only induces an axial stress but also tangential and radial stresses. The tangential and radial stresses have contributions to the strain in axial direction. More important, the actual axial stress in the cylinder is not uniform and cannot be represented by simply dividing the applied force with the cross-section area of the cylinder. Therefore, the slope of the stress-strain curve in Fig. 10 does not give directly the transverse compressive E-modulus. Using the finite element method or the analytical formula in [6], we can calculate the distribution of the axial, tangential and radial stresses as shown in Fig. 11. The axial strain ε$_z$ in relation to the applied axial force *F* is expressed as the following:

$$e_z = \frac{F}{E_z p b^2}\left\{(1-c^2)+2h\left[\frac{(1-c^2)(n_{rz}+n_{rq})}{2}+\frac{(1-c^{(g+1)})^2(n_{rz}+gn_{rq})}{(g+1)(1-c^{2g})}-\frac{(1-c^{(g-1)})^2c^2(n_{rz}-gn_{rq})}{(g-1)(1-c^{2g})}\right]\right\}^{-1} \quad (2)$$

where $h=\frac{(n_{rz}-n_{rq})E_q}{(1-n_{rz}^2-n_{rq}^2)E_q-E_z}$, $g=\sqrt{\frac{(1-n_{rz}^2)E_q}{(1-n_{qr}n_{rq})E_z}}$, and $c=\frac{a}{b}$, where *a* and *b* are the inner and outer radius of the cylinder, respectively. The transverse compressive E-modulus is found to be 3 GPa for the sample shown in Fig. 10. This is about the same as that found in bar-shaped samples.

The matrix material used in the experiment is epoxy (Stycast 1266). The compressive stress-strain curve of pure epoxy is shown in Fig. 12. The elastic modulus of the epoxy is about 3.1 GPa, the same as that found in the tensile test. Therefore, the low transverse elastic modulus of the Zylon/epoxy composite should originate from the low transverse elastic modulus of Zylon fibre itself according to the rule of mixture [4] or caused by structural problems.

The PBO molecules are highly oriented in the micro-fibrils (orientation factor > 0.95) in a Zylon fibre. However, there are micro-voids between the micro-fibrils along the fibre direction. The length of a void is about 50 nm and the mean void diameter is approximately 3 nm, compared to the micro-fibril's diameter of 10 to 50 nm. Due to the presence of the micro-voids in the fibre, it is possible that the fibre deforms easier in the transverse direction than in longitudinal direction and hence shows the low transverse E-modulus. This is also in agreement with the observation that the fibres deform to a hexagonal shape when pressed against each other in the Zylon/epoxy samples with very high filling factor (>90%), as mentioned before.

The ultimate compressive strength of the Zylon/epoxy composite samples is also very low, only 50 to 150 MPa. The failure of a cylindrical sample occurs in the inner layers (about 1/3 of the wall-thickness). The fibres in these layers buckle into the inner hole. A bar-shaped sample cracks along a plane parallel to the fibre and about 30° from the loading direction (Fig. 13). The cracks start from the corner. The close packed fibre structure has a tendency to slide along a plane 30° from the loading direction when the force components are not perfectly in balance. Furthermore, the circular cross-section of the fibre introduces a force perpendicular to the applied compressive force. This "billiard ball" effect will weaken the structure if no constraint is present in the boundary. In a cylindrical sample, the effect is restricted more efficiently by the outer layers of the fibres. Therefore, the cylindrical sample shows a higher ultimate compressive strength than the bar-shaped sample.

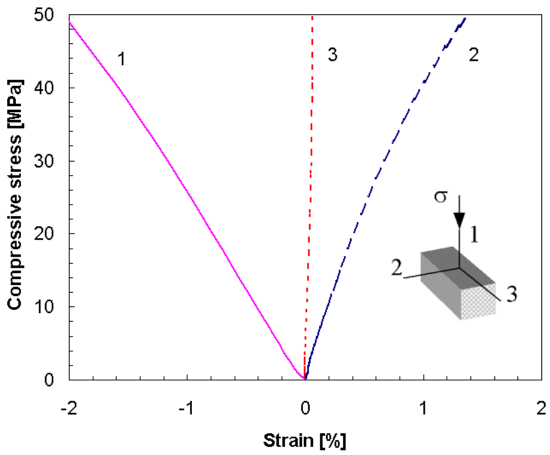


Fig. 9 Transverse compressive stress-strain curve of a bar-shaped sample of Zylon/epoxy composite (filling factor = 77.5%). The fibres are along direction 3.

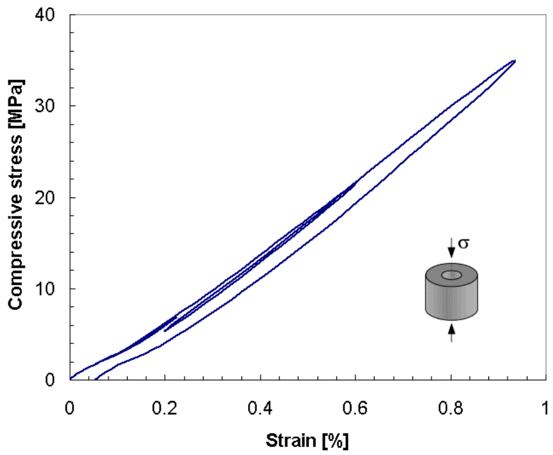


Fig. 10 Transverse compressive stress-strain curve of a cylindrical sample of Zylon/epoxy composite (filling factor = 81.4%). The stress cycling causes hysteresis loops in the curve.

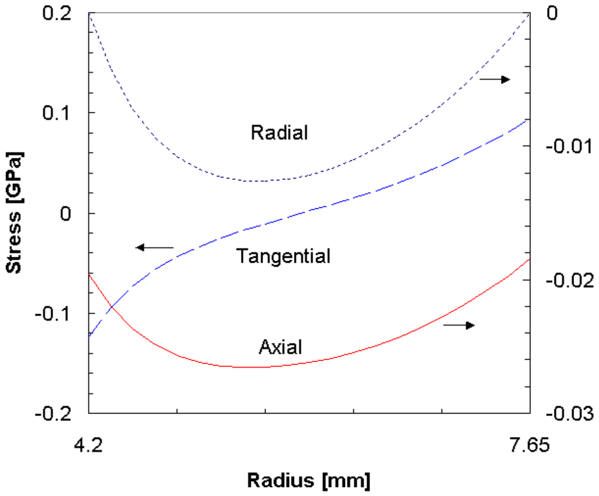


Fig. 11 The calculated distribution of axial, tangential and radial stresses in the cylindrical sample (Fig. 10) under uni-axial compressive force (3.08 kN). The axial strain is 0.6%.

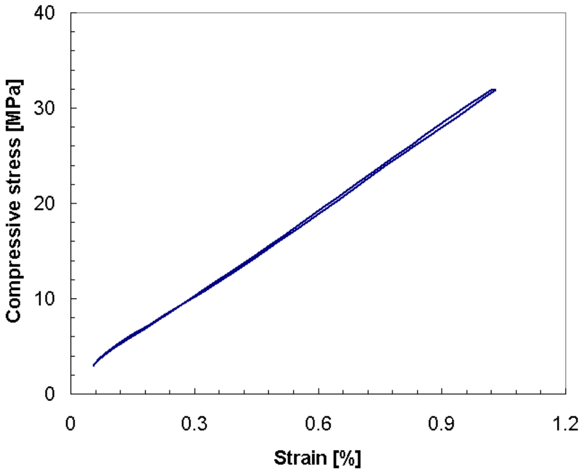


Fig. 12 Compressive stress-strain curve of pure epoxy (Stycast 1266).

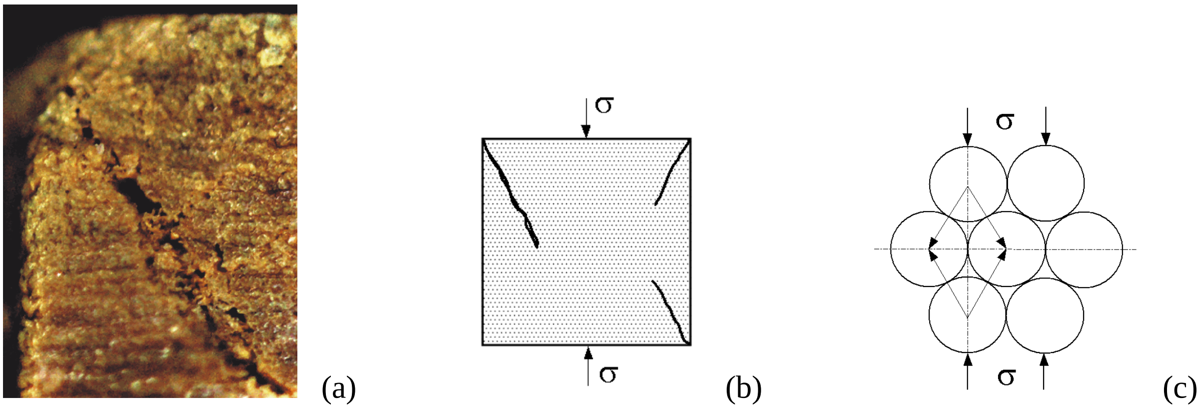


Fig. 13 (a) Crack resulting from the transverse compressive stress in a bar-shaped sample; (b) The schematic cross-section showing the positions of the cracks and the crack development; (c) The microscopic fibre-structure showing the tendency of slip along a plane 30° from the loading direction when the force components are not perfectly in balance.

4. Conclusions

The Zylon/epoxy composite with a high fibre filling factor can be easily fabricated by wet-winding the Zylon fibre with epoxy (Stycast 1266). The pre-stress applied to the Zylon fibre bundle during winding is necessary for obtaining the high filling factor and homogenous distribution of fibre in the matrix. The ultimate tensile strength (UTS) of the composite is mainly determined by the fraction of the Zylon fibre. The UTS of the Zylon fibre in the composite is found to be 4.3 GPa at room temperature and is about 30% higher at 77 K. These values may be affected by the pre-stress conditions. The Zylon/epoxy composite shows a very low transverse compressive elastic modulus and ultimate compressive strength (UCS). This is attributed to the microstructure of the fibre itself and the fibre configuration in the composite. The mechanical properties of Zylon/epoxy composite are summarised in Table 2. In view of the structure and the stress state of a magnet, the fibre-wound Zylon/epoxy composite is suitable as reinforcement material for the high-field magnet coils because of its very high UTS along the fibre direction.

The fabrication process of the composite in cylindrical shells is easy to implement in magnet coil winding.

Table 2 Mechanical properties of Zylon/epoxy composites. The data are normalised to a filling factor of 77.5%.

Temp.	Uni-axial tensile tests (along fibre)			Transverse compressive tests			
[K]	UTS [GPa]	E_3 [GPa]	ν_{31}	UCS [MPa]	E_1 [GPa]	ν_{12}	ν_{13}
R.T.	3.3	205	0.35	60~150	3	0.6	~0.005*
77	4.3	222	0.35				

*calculated with Eq. (1).

Acknowledgements

The authors would like to thank Dr. P.J. Nederveen of TU Delft for helpful discussions.

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