Flexural Characterisation of Unidirectional Thermoplastic Tapes using a Dynamic Mechanical Analysis system

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Abstract
The characterisation of accurate material properties is a prerequisite for any forming simulation. Due to their characteristics, the mechanical behaviour of pre-impregnated Carbon Fibre Reinforced Polymers must be investigated under controlled environmental conditions, i.e. humidity and temperature. In this work, flexural properties in fibre direction of unidirectional thermoplastic tapes (PA6) are investigated at different temperatures using a Dynamic Mechanical Analysis system. In a first step, four different types of flexural setups are assessed (cantilevers, 3-point-bendings) with specimens of two different thicknesses at three temperatures and three loading rates. Once the most suitable test setup-specimen combination is defined, quasistatic tests are conducted in order to assess temperature and loading rate dependencies of the material.

1. Introduction

Presenting high specific stiffness pre-impregnated Carbon Fibre Reinforced Polymer (CFRP) materials have been used for a long time in the manufacturing of composite components [1]. The demand of high performance/high temperature thermoplastic composites is expected to grow in the next years because of their interesting inherent properties [2]. A common method to process pre-impregnated thermoplastic composites is to use a thermoforming press. The flat composite layup is placed on top of a mould, heated above melting temperature and formed. The combination of pressure and a high temperature, which softens the material, enables the material to deform to a specific geometry.

In order to predict the outcome of a thermoforming process before any experimentation, forming simulations using Finite Element (FE) methods are recommended. Although forming simulations of dry materials, i.e. non-crimp fabrics, woven fabrics, have been mainly developed so far [3, 4], forming simulations of pre-impregnated materials tend to be more investigated these days [5, 6, 7]. Achieving meaningful results by FE models is only possible if accurate material properties are available. These must be defined under the same environmental conditions as the forming occurs. Material characterisation of pre-impregnated materials must thus be conducted at high temperature.

Typical investigations concern properties in tension, bending and shear [8]. The characterisation of pre-impregnated materials is commonly performed on adapted versions of tests used for dry reinforcements at room temperature. For example, the shear behaviour of pre-impregnated composites is often sought with picture frame or bias-extension tests within a thermal chamber mounted on a tensile testing machine [9, 10]. However, such adjustments are not always possible. In the case of flexural investigations, current test methods built on the cantilever test introduced by Peirce cannot be adapted [11]. Therefore, new approaches were developed using a universal testing machine, a regulated thermal chamber and three- as well as four-point bending tests [12, 13]. A new approach using a Dynamic Mechanical Analysis (DMA) system, which allows a better control of the testing parameters, is presented in this paper.

2. Experimental method

2.1. Material

The material, provided by Ticona and referenced to as Celstran®, is a commercially available continuous unidirectional carbon fibre reinforced thermoplastic composite tape made out of polyamide 6 (PA6). PA6 is a semi-crystalline thermoplastic. As such, it has the particularity to remain solid until its melting temperature. Afterwards, it becomes highly viscous and behaves like viscoelastic fluids [14]. All properties presented in Table 1 are provided by the material supplier, except for the melting temperature that was measured using a Differential Scanning Calorimetry system (DSC Q2000, TA Instruments).

Density [g.cm^{-3}]	**Tape thickness [mm]**	**Glass transition temperature [°C]**	**Melting temperature (measured) [°C]**
1.46	0.19	47	219

Table 1. Main properties of unidirectional PA6 Tapes (Celstran®, Ticona)

2.2. Equipment

Within this study, a Q800 machine from TA Instruments is used. Humidity conditions are not controlled. The machine is placed in a laboratory with an ambient temperature of 23°C. The Q800 enables fine measurements over an extended range of temperatures (-145°C to +600°C). Heating rates from 0.1 up to 20°.min^{-1} can be applied. Based on the information provided by the supplier, the temperature reproducibility is ±2°C. The machine can test samples of stiffness between 10^2 and 10^7 N.m^{-1}. [15]

Several different types of clamps can be adapted to the DMA system, i.e. tension, compression and bending, and either used in dynamic or quasistatic modes [15, 16]. In the scope of this work, only quasistatic tests are considered.

2.3. Specimen preparation

The PA6 tapes were laid down using a Fiberforge® Station and then consolidated using a thermoforming press.

The fibre orientation on the surface of the specimens was controlled using a ProFactor® sensor. This verification ensures that the samples have a homogeneous unidirectional fibre direction parallel to the side length of the specimens. This aspect is important in order to avoid any off-axis loading while testing.

Given that four different test setups (single cantilever, double cantilever, three-point bending 20 mm and three-point bending 50 mm) and two sample geometries (specimens of 1 mm and 2 mm thickness) are assessed, eight sorts of samples had to be prepared. While the 2 mm specimens consisted of 14 unidirectional plies, the 1 mm specimens had only 7 plies. The approximated dimensions of the specimens are given in Table 2.

	Single Cantilever	**Double Cantilever**	**Three-point bending (20mm)**	**Three-point bending (50mm)**
Length [mm]	17.5	35	20	50
Width [mm]	15	15	15	15
Thickness [mm]	1and 2	1and 2	1and 2	1and 2

Table 2. Dimensions of the specimens for the different bending tests

Specimens were stored at room temperature without any special conditioning. Given that PA6 is hygroscopic, i.e. subjected to water absorption [1], the specimens were dried at 75°C for 14 hours prior to testing. Even though the exact water content within the samples could not be precisely determined, it can be considered as reasonably low [17].

2.4. Test series

2.4.1 Description

As a start, investigations were performed to determine the most suitable test setup-specimen combination for the determination of flexural properties of PA6 tapes. Tests were conducted at 200°C, 230°C and 260°C, i.e. far below, above, and far above melting temperature, respectively. Three loading rates were applied: 10 µm.min^{-1}, 100 µm.min^{-1} and 500 µm.min^{-1}. Due to the stiffness limit of the machine, three-point bending (50 mm) tests with specimens of 1 mm thickness could not be conducted.

Afterwards, tests were carried out at different temperatures (from 200°C to 280°C) and different loading rates (from 10 µm.min^{-1} to 10000 µm.min^{-1}).

2.4.2. Procedure

The specimens were positioned in the machine. The alignment of the sample was appropriate when the width of the specimens was parallel to the clamps/support. A good position of the sample is essential to avoid unsymmetrical loadings. Specimens mounted on different test setups are shown in Figure 1.

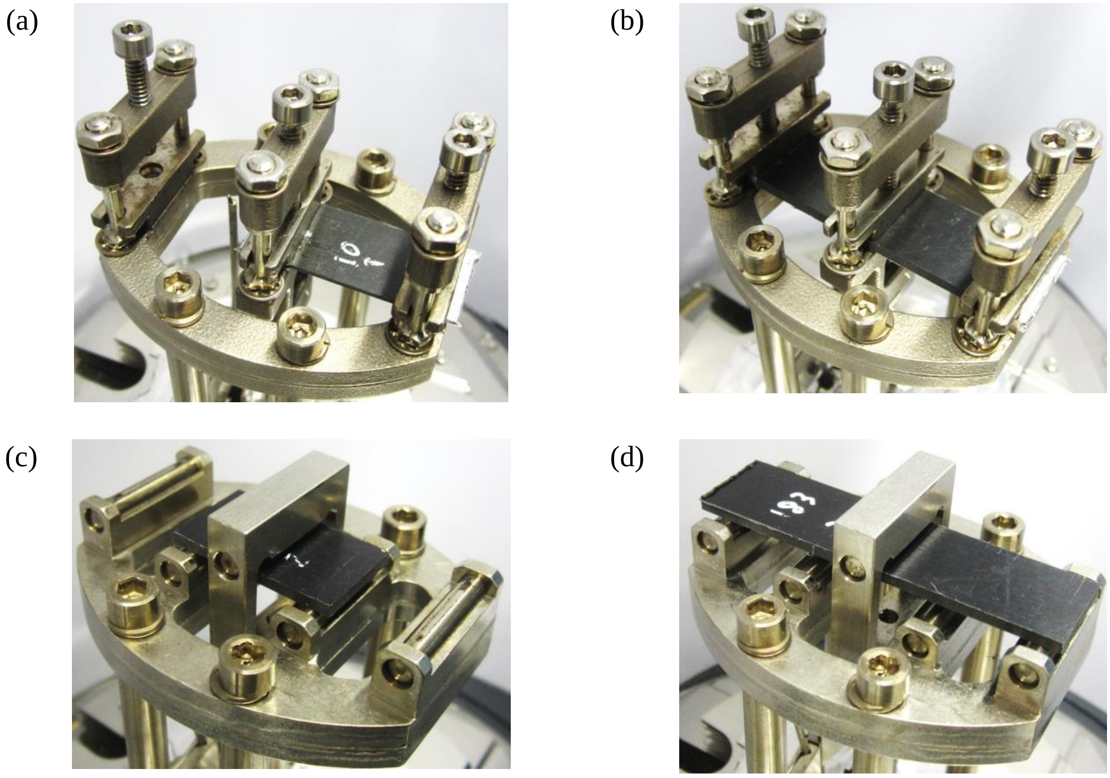

Figure 1. Specimens mounted on different test setups. (a) Single cantilever, (b) Double cantilever, (c) Three-point bending (20 mm), (d) Three-point bending (50 mm)

Tests conducted with the single/double cantilever setups were clamped with a torque of 0.79 N.m (7 in.lb). Clamps were tightened at room temperature and retightened using the same torque at 210°C for tests at 230°C and 260°C to ensure a proper contact between the composite and the tooling. Regarding three-point bending tests, a preload of 0.001 N was applied to the specimen. Then, the furnace was closed and the specimen was heated up to the testing temperature (200°C, 230°C or 260°C) either at a heating rate of 10°.min^{-1} or 20°.min^{-1}. To ensure a homogeneous temperature distribution within the specimen, tests were performed five minutes after isothermal conditions were reached. Finally, tests were executed with controlled displacement ramps in order to control the loading rate.

3. Results and discussion

3.1. Flexural test selection

The first test series focused on the selection of the most suitable test setup-specimen combination for the investigation of flexural properties of PA6 tapes.

It was noticed that both cantilever tests (single and double) were suitable for testing below melting temperature, i.e. 200°C, but not above, i.e. 230°C and 260°C. Above melting temperature, both test setups appeared to be instable. Their instability was caused by the lack of reactivity of the machine. The DMA system did not manage to take the transition of the material into account and adjust its parameters accordingly. As a consequence, when the melting temperature was reached, the sample was damaged due to the application of high forces. This issue was overcome by forcing the machine to stay isothermal below melting temperature, e.g. 215°C, for long period of time. Accordingly this method does not allow rapid testing. Besides, it is reported in literature that the clamping of the cantilever tests has an undesirable effect on the outcome [18, 19, 20]. Therefore, cantilever tests are not suitable for this work.

Tests with the three-point bending (50 mm) setup appeared to be stable. However, due to the tooling arrangement – both three-point bending setups (20 mm and 50 mm) are built up in the same rig – this test setup could not be selected (see Figure 1 (c)). In case of high displacements, the specimen could come into contact with the three-point bending (20 mm) tooling. This is not acceptable and therefore this test is not suitable for this work.

Testing with the three-point bending (20 mm) setup was successful. Nevertheless, only specimens of 1 mm thickness were suitable.

		Width before testing [mm]	**Width after testing [mm]**	**Variation [%]**
1mm specimen	**500 $\mu m.min^{-1}$**	14,85	15,21	+2,42
	100 $\mu m.min^{-1}$	15,00	15,40	+2,67
	10 $\mu m.min^{-1}$	13,90	13,93	+0,22
2mm specimen	**500 $\mu m.min^{-1}$**	14,66	17,27	+17,80
	100 $\mu m.min^{-1}$	14,65	16,99	+15,97
	10 $\mu m.min^{-1}$	14,73	16,42	+11,47

Table 3. Three-point bending (20 mm) setup: variation in specimen widths at 230°C

As it can be seen in Table 3, the width of the specimens of 2 mm thickness was significantly modified by the testing. Thickness measurements were not possible due to the deformed shape of the specimens. Similar deformations were also obtained at 260°C.

The ratio of 20:1 between free length span and specimen thickness also supports the choice of using specimens of 1 mm thickness. According to the norm ISO 14125, using such a ratio minimises the occurrence of inter-laminar shear within the sample [13].

The results of this first test series tend to consider the three-point bending (20 mm) test setup and specimens of 1 mm thickness for further investigations.

3.2. Characterisation at forming temperature

With the selected test method, i.e. three-point bending (20 mm) and specimens of 1 mm thickness, PA6 tapes were characterised. In a first step, the dependency to temperature was investigated (see Figure 2).

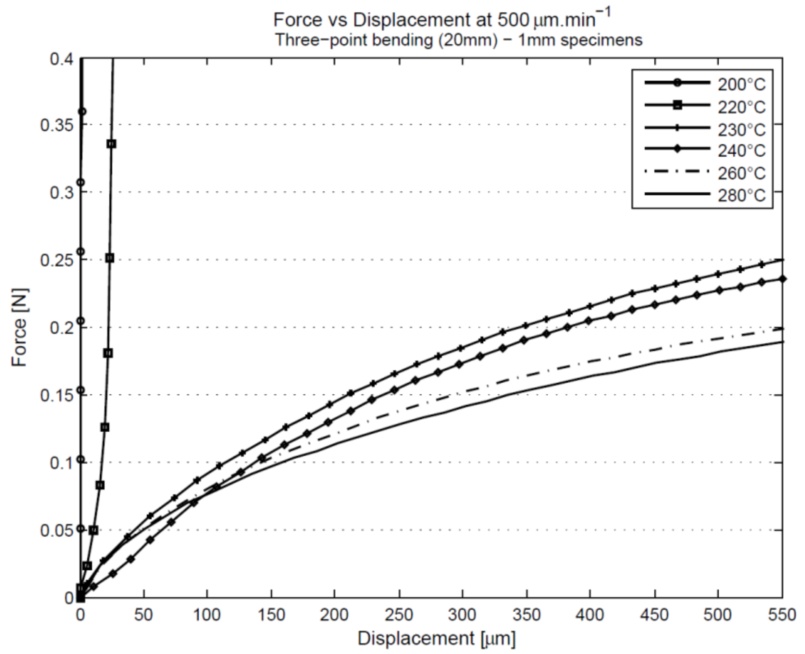

Figure 2. Effect of temperature on three-point bending tests (20 mm) at 500 µm.min^{-1} with 1 mm specimens

Subsequently, PA6 tapes were characterised at forming temperature (280°C) for different loading rates (see Figure 3) in order to investigate the viscoelastic behaviour of the material.

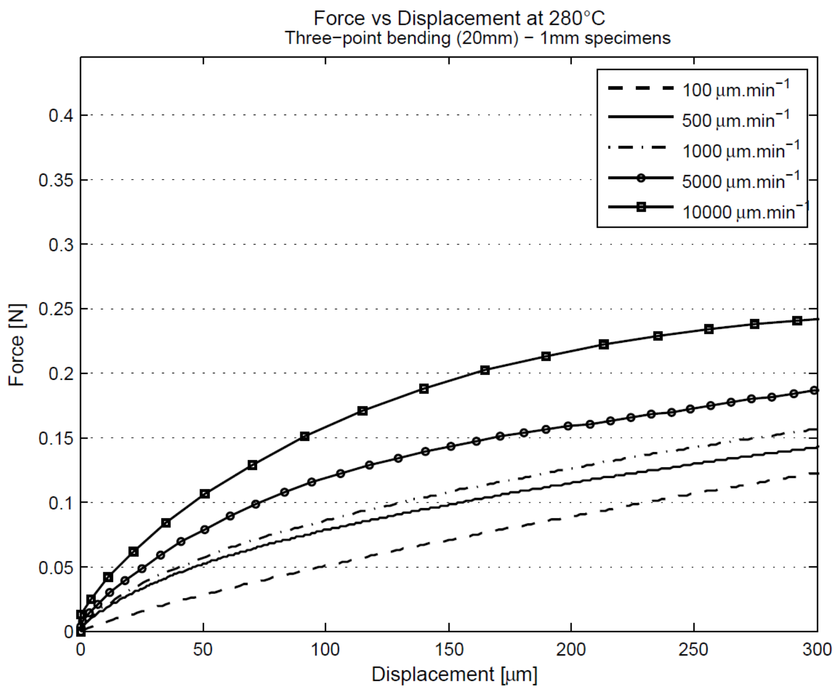

Figure 3. Effect of loading rate on three-point bending tests (20 mm) with 1 mm specimens

As it can be seen in Figure 2, as the temperature increases, forces decrease. The weak early behaviour of the curve at 240°C is due to the acceleration of the moving clamp of the DMA system. The requested loading rate of 500 $\mu m.min^{-1}$ was not applied directly at the beginning of the test but was reached after a displacement of 50 µm. When the loading rate is stabilised, i.e. after about a displacement of 200 µm, results are in compliance with the expectations [21].

As it can be seen in Figure 3, as the loading rate increases, forces increase. This is a typical behaviour of semi-crystalline thermoplastic composite materials above their melting temperature [21]. It must be noted that the application of the loading rate may be inaccurate at the beginning of the test due to the acceleration of the moving clamp of the DMA system. This effect is amplified by high loading rates.

4. Conclusion

The investigation of flexural properties in fibre direction of PA6 tapes using a Dynamic Mechanical Analysis system was presented within this work. In a first step, two different types of specimens (1 mm and 2 mm thickness) were assessed with four different types of flexural setups (single/double cantilever, three-point bending 20 mm and three-point bending 50 mm). It was shown that testing with a three-point bending (20 mm) setup and specimens of 1 mm thickness is the most promising combination. Therefore, it was used to characterise the material. Results showed important temperature and loading rate dependencies.

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References

[1] B.T. Ålström. *Manufacturing of Polymer Composites.* Chapman & Hall. London, 1997.
[2] High-temp thermoplastics: Higher Expectations. http://www.compositesworld.com/articles/high-temp-thermoplastics-higher-expectations. Composites World, 2012.
[3] P. Boisse, N. Hamila, F. Helenon, B. Hagege, J. Cao. Different approaches for woven composite reinforcement forming simulation. *International Journal of Material Forming*. 1:21–29, 2008.
[4] K.A. Fetfatsidis, J.A. Sherwood. Simulation of the Manufacturing of Non-Crimp Fabric-Reinforced Composite Wind Turbine Blades to Predict the Formation of Wave Defects. In *ESAFORM*. Volume 1353, 895-900, 2011.
[5] P. Wang, N. Hamila, P. Boisse. Thermoforming simulation of multilayer composites with continuous fibres and thermoplastic matrix. *Composites: Part B.* 52:127–136, 2013.

[6] P. Harrison, R. Gomas, N. Currado-Correia. Press forming a 0/90 cross-ply advanced thermoplastic composite using the double-dome benchmark geometry. *Composites: Part A*, 54:56-69,2013.
[7] S.P. Haanappel, R.H.W. ten Thije, U. Sachs, B. Rietman, R. Akkerman, Formability analyses of uni-directional and textile reinforced thermoplastics. *Composites : Part A.* 56 :80–92, 2014.
[8] P. Boisse, N. Hamila, E. Vidal-Sallé and F. Dumont. Simulation of wrinkling during textile composite reinforcement forming. Influence of tensile, in-plane shear and bending stiffnesses. *Composites: Part A*, 71(5):683–692, 2011.
[9] P. Harrison, M.J. Clifford, A.C. Long. Shear characterisation of viscous woven textile composites: a comparison between picture frame and bias extension experiments. *Composite Science and Technology*, 64(10–11):1453–1465, 2004.
[10] J. Cao et al. *Characterization of mechanical behavior of woven fabrics: Experimental methods and benchmark results*. Composites: Part A, 39:1037–1053, 2008.
[11] F.T. Peirce. The "Handle" of Cloth as a Measurable Quantity. *Journal of the Textile Institute Transactions*. 21:377-416, 1930.
[12] ASTM D790. Testing the flexural properties of Plastics and Polymers, 1993.
[13] Norm DIN EN ISO 14125. Fibre-reinforced plastic composites - Determination of flexural properties, 1998.
[14] T.A. Martin, D. Bhattacharyya, *Squeeze flow in thermoplastic composites*. In Polypropylene: An A-Z reference; Polymer Science and technology Series Volume 2, 776-782, 1999.
[15] TA Instruments. DMA Q Series™ Getting Started Guide, Revision H, 2007.
[16] K. P. Menard. *Dynamic mechanical analysis: a practical introduction.* CRC Press LLC, Boca Raton, 1999.
[17] Suherman, M. Peglow, E. Tsotsas. Drying kinetics of granular Nylon-6. In *European Congress of Chemical Engineering (ECCE-6)*, Poster, 2007.
[18] S. Deng, M. Hou, L. Ye. *Temperature-dependent elastic moduli of epoxies measured by DMA and their correlations to mechanical testing data*. Polymer Testing, 26:803–813, 2007.
[19] S.J. Hwang, R.F. Gibson, J. Singh. Decomposition of coupling effects on damping of laminated composites under flexural vibration. *Composites Science and Technology*, 43:159–69, 1992.
[20] J.D.D. Melo, D.W. Radford. Time and temperature dependence of the viscoelastic properties of CFRP by dynamic mechanical analysis. *Composite Structures*, 70:240–253, 2005.
[21] W. Grellmann, S. Seidler. *Polymer Testing*. Carl Hanser Verlag GmbH & Co. KG, München, 2007.