INVESTIGATION OF MECHANICAL PROPERTIES OF NONWOVEN SECOND GENERATION COMPOSITE MATERIAL ELABORATED THROUGH A MIXTURE OF CARBON FIBERS AND FILAMENT LENGTHS

Sabrine Jlassi[1], Florentin Berthet[1], Gérard Bernhart[1]

[1] Institut Clément Ader (ICA), Université de Toulouse, CNRS, IMT Mines Albi, INSA, ISAE-SUPAERO, UPS, Campus Jarlard, 81013 Albi CT Cedex 09, France
Email: sabrine.jlassi@mines-albi.fr, florentin.berthet@mines-albi.fr, gerard.bernhart@ mines-albi.fr,
Web Page: http://institut-clement-ader.org/

Keywords: carbon fiber, recycling, steam thermolysis process, thermoplastic composites, nonwovens.

Abstract

The current study aims to investigate the mechanical properties of ten different carbon fiber nonwoven reinforced thermoplastic composites. A design of experiments was carried out by using a Mixture Design methodology in order to produce nonwoven fabrics by carding and pre-needling. Composites were manufactured by compression molding using film stacking method. Tensile properties such as tensile modulus and tensile strength were investigated as function of proportion of fibers in nonwovens and direction of applied load. It was found that the mechanical properties of the composites were much higher in the Cross direction than in the Machine direction. It was observed that the mechanical properties are mainly influenced by fiber properties and orientation of fibers in the nonwovens. The tensile modulus was then modelled by multilinear regression resulting from the design analysis and Rule of Hybrid Mixtures (RoHMs). A good correlation between the experimental and the predicted values is observed.

1. Introduction

During the last few years, there has been great deal of interest in recycling carbon-fiber-reinforced polymer composites (CFRP). Different technologies have been proposed to treat CFRP and recover carbon fiber for reuse in 2nd generation Composites [1-2]. The steam thermolysis process is a thermo-chemical recycling process using superheated steam water to degrade organic matrix and recover carbon fibers from CFRP [3]. It was shown that the reclaimed carbon fibers (rCF) have retained their original mechanical properties [4].

From an industrial and economical point of view, there is a major challenge related to the sorting of composites by type/grade of fibers before recycling. An optimal solution would be to be able to produce rCF nonwovens with guarantied properties avoiding the initial CFRP sorting step and to make cost effective 2nd generation composites. Many studies have been carried out on the effect of rCF properties as function of recycling process on the mechanical performance of composites [2–5]. Small number of studies have been reported the influence of variability of sources of rCF on the mechanical properties of composites. Actually, the fibers are mixed during the recycling process, so that reclaimed fibers present a distribution of properties [1]. In this case, ELG CF explained that blending different products reduces property variation in second generation products [6].

This study aims to investigate the influence of carbon fiber sources variability on the mechanical properties of nonwoven reinforced composites. A design of experiments was carried out by using a Mixture Design methodology in order to produce different nonwoven fabrics [7]. Three different virgin carbon fiber tows (T300, T700 and IM7) were selected and cut into lengths up to 110 mm. According to the mixture design, ten nonwoven fabrics, with a weight close to 200g/m², were prepared by carding and needle punching (pre-needling) using the same process parameters. These nonwovens were associated to Polyamide 6 (PA6) matrix films to produce by compression molding process ten composites with a fiber volume fraction of 28 vol%. Composite mechanical properties were evaluated by performing tensile test along the machine direction (MD), which is the direction of nonwoven production, and the cross direction (CD).

2. Materials and methods

2.1. Materials

In this study three virgin carbon fiber tows, T300, T700 (Toray) and IM7 (Hexcel), were selected to produce nonwovens. The basic properties of these fibers are presented in Table 1. They represent high strength and intermediate modulus-high strength carbon fibers. The selection was based on the mechanical properties of the fibers and their use in aircraft and automotive applications (end-of-life composites are generally reinforced by these fibers). The fibers were cut into three lengths 50 mm, 80mm and 110mm.

A design of experiments was carried out by using a Mixture Design methodology in order to produce nonwovens and investigate the mechanical properties of nonwoven reinforced composites (Table 2). The Figure 1 shows a diagram of the design with 10 experiments. Each corner of the diagram consists of 100% of one of the three fibers cut into different lengths. The sides represent binary mixtures. Generally, the first 7 points are used to estimate the regression model, the 3 following to validate it.

The nonwovens were produced via a carding/cross lapping/needle-punching process at STFI (Saxon Textile Research Institute e.V.) in Germany. The fibers were mixed according to the mixture design and carded to be transformed into a veil. The carded veils were then cross lapped and pre-needled (single sided needling, 60 punches/cm², 14mm needle depth), in order to form 100% carbon fiber nonwovens with an areal density of 200g/m².

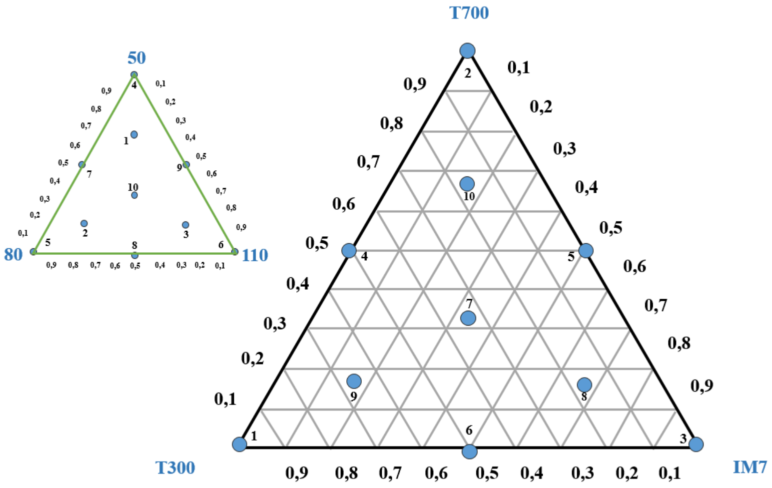


Figure 1. A Simplex centroid design for a three different carbon fiber and fiber length mixture.

Table 1. Structural and mechanical properties of material constituents.

Fiber/Matrix	Density (g/cm^3)	Tensile strength (MPa)	Tensile Modulus (GPa)	Elongation (%)	Length (mm)
T300	1.76	3530	230	1.5	50 / 80 / 110
T700	1.8	4900	230	2.1	50 / 80 / 110
IM7	1.78	5760	290	2.0	50 / 80 / 110
PA6	1.14	90	3.2	4.5	---

Table 2. Design of mixture table

N°	Fiber			Fiber length (mm)		
	T300	T700	IM7	50	80	110
1	1	0	0	0,1665	0,667	0,1665
2	0	1	0	0,1665	0,165	0,667
3	0	0	1	0,667	0,1665	0,1665
4	0,5	0,5	0	1	0	0
5	0	0,5	0,5	0	1	0
6	0,5	0	0,5	0	0	1
7	0,333	0,333	0,333	0,5	0,5	0
8	0,1665	0,667	0,1665	0	0,5	0,5
9	0,1665	0,165	0,667	0,5	0	0,5
10	0,667	0,1665	0,1665	0,333	0,333	0,333

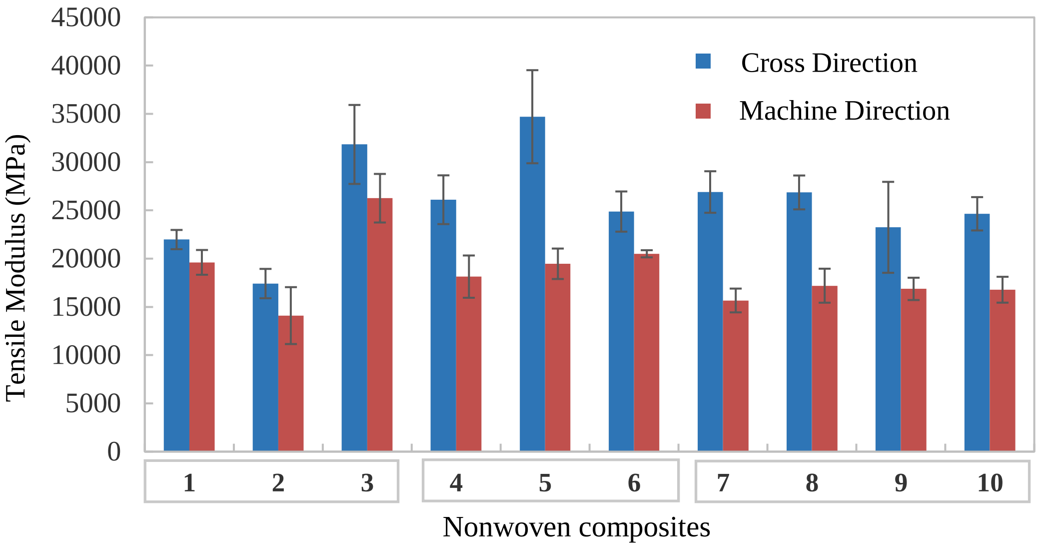
2.2. Composite preparation and testing method

Composite plates with 28% fiber volume content and 2 mm thickness were manufactured by compression molding using film stacking method. In this method, carbon fiber nonwovens and PA6 films were stacked alternately. Before manufacturing, the PA6 films were dried in an oven at 80°C for 24 hours. The obtained composite plates were cut into tensile specimens in accordance with ISO-527 specification with dimensions 250 mm x 25mm x 2mm. Specimen ends were reinforced by gluing ±45°glass fiber/epoxy end tabs to avoid sample failure out of the gage length. Tensile properties were determined according to the ISO 527 standard using 30 KN INSTRON-5200 testing machine at a crosshead speed of 1 mm/min. An extensometer was used to record strain of tested specimens. Composite mechanical properties were evaluated by performing tensile tests along the machine direction (MD) and the cross direction (CD).

3. Results and discussion

3.1. Experimental results

The mechanical properties of the ten composites obtained from tensile tests are summarized in cluster column charts shown in Figure 2 and Figure 3. Each test data chart is the average value from three to five identical specimens and the error bars on the histograms represent the standard deviation. These results show that the tensile modulus and strength values range respectively from 18 GPa to 35 GPa and 200 MPa to 550 MPa in the CD direction. In the MD direction the values range from 15 GPa to 25 GPa for the modulus and 150 MPa to 450 MPa for strength. It is important to note that the results are different depending on the direction of applied load. The composites have higher mechanical properties in the CD direction. This could be attributed to the fiber orientation distribution in nonwovens which could be influenced by the fiber lengths and the process parameters. Indeed, in carded and needle-punched nonwovens, the preferential orientation of fibers is close to the CD direction. This phenomenon explains the anisotropy of the mechanical properties of nonwoven composites.



Error Bars: ± Standard Deviation

Figure 2. Tensile modulus of nonwoven reinforced composites in machine and cross directions.

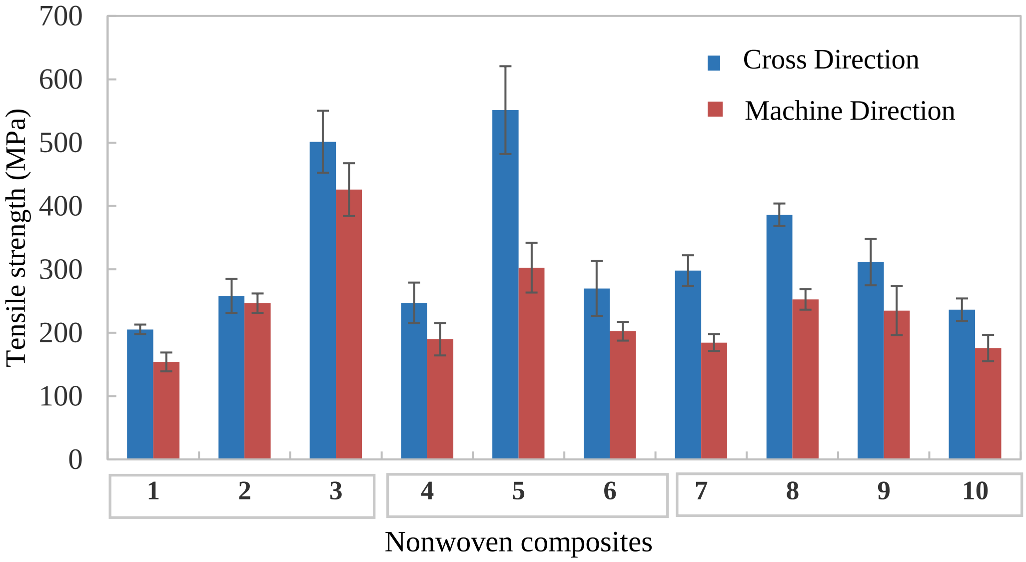


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Figure 3. Tensile strength of nonwoven reinforced composites in machine and cross directions.

According to the Table 2, the experimental design can be divided into 3 groups. The first group corresponds to the composites C1, C2 and C3. These composites are reinforced by 100% of each fiber cut into different lengths. Due to the best mechanical properties of IM7 fiber, C3 presents the highest modulus and strength in both MD and CD directions. The second group corresponds to the composites C4, C5 and C6. In this case, composites are reinforced by a mixture of two fibers (50/50) cut into one fixed length. The best properties obtained for the C5 in the CD direction. For this composite the cut length fixed at 80 mm and the presence of IM7 fiber in the mixture have increased the modulus as well as the resistance in this direction. The last group corresponds to the rest of the design of experiments (C7, C8, C9 and C10). The composites were reinforced by all the fibers cut into three different lengths.

No significant difference was observed between the tensile properties of the composites in both MD and CD directions.

In addition to these investigations, an analysis of fiber orientation in nonwovens was carried out in order to predict the tensile modulus of nonwoven reinforced composites. A regression model was used to predict the contribution of fibers on the tensile modulus. Moreover, a modified Rule of Hybrid Mixtures was developed to predict the composite tensile modulus. This model takes into account the fiber orientation distribution and tensile properties.

3.2 Theoretical Analysis: Tensile modulus prediction

In order to study the influence of fiber properties and architecture on the tensile modulus of the composites in MD and CD directions, a regression model and Rule of Hybrid Mixtures were used.

- **Regression model** [5]

A statistical regression model considering fiber proportions as variables and tensile modulus as the response produced the regression line. In this study, tensile modulus be evaluated by the regression equation as follows:

$$Y = A\,X_{T300} + B\,X_{T700} + C\,X_{IM7}. \tag{1}$$

Where A, B and C are the regression coefficients; X_i is the proportion of each fiber “i” in the composite. Typically, the coefficients have to be tested as significantly different from “0” or not. This can be done by evaluating the probability values which have to be less than 5% [5].
Modulus in CD and MD can be evaluated by the regression equations (2) and (3). In this case the different coefficients are significant and the squared multiple correlation R^2 is equal to 0.94 for CD regression and 0.95 for MD regression.

$$E_c\,(CD) = 22863\,X_{T300} + 23581\,X_{T700} + 32216\,X_{IM7}. \tag{2}$$

$$E_c\,(MD) = 18068\,X_{T300} + 14097\,X_{T700} + 22624\,X_{IM7}. \tag{3}$$

- **Rule of Hybrid Mixtures RoHMs** [8-9-10]

In this study, by mixing different carbon fibers, the composites can be assimilated as hybrid materials. Several studies have been carried out on the effect of carbon fiber hybrid with glass fibers [8-9]. But, some research has been reported the effect of hybrid composites reinforced with more than two different carbon fibers. In this case the tensile modulus of hybrid nonwoven composite can be predicted using equation 4.

$$E_{hc} = E_{T300}\,V_{T300} + E_{T700}\,V_{T700} + E_{IM7}\,V_{IM7}. \tag{4}$$

Where E_{hc} is the tensile modulus of hybrid composite and V_{T300}, V_{T700} and V_{IM7} are the proportion of each fiber in the composite. E_{T300}, E_{T700} and E_{IM7} are respectively tensile modulus of T300/PA6, T700/PA6 and IM7/PA6 composites with 28% fiber volume fraction and can be predicted using modified rule of mixture (equation 5) developed by Cox [11] and modified by Krenchel [12].

$$E_i = \eta_o\,E_f\,Vf + E_m\,(1-V_f). \tag{5}$$

$$\eta_o = \Sigma\,a_{(i)}\cos^4\theta_{(i)}\,. \tag{6}$$

Where E_f and E_m are tensile modulus of fiber and matrix, V_f is the fiber volume content and η_o is an efficiency factor which takes into account the proportion of fibers $a_{(i)}$ oriented according to $\theta_{(i)}$ in the nonwoven fabrics. This factor was determined using image analysis method and the technique used is known as Fast Fourier Transform (FFT) [13]. The analyzed images correspond to the SEM observations of nonwoven fabrics as shown in Figure 4. The orientation factor was then determined from the polar plot of fiber orientation distribution using Equation 6. It is found that values of η_o for all the different nonwoven fabrics studied are close to each other. The obtained η_o in MD and CD directions are equal respectively to 0.266 and 0.464.

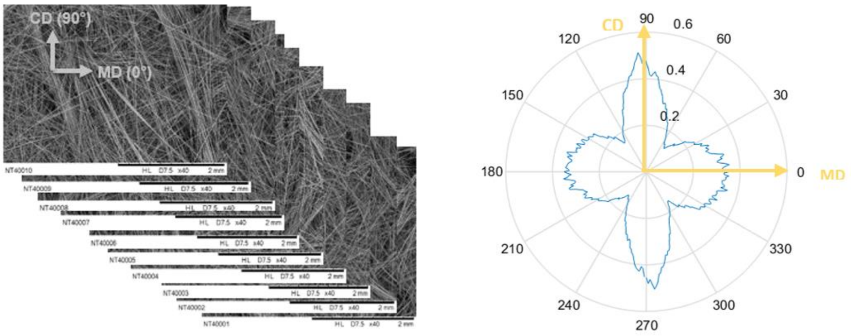
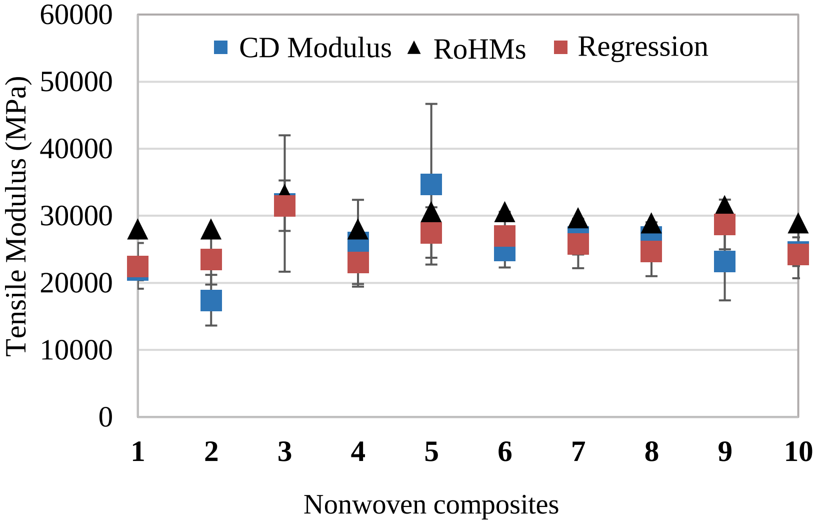


Figure 4. Polar plot of fiber orientation distribution in nonwoven fabric.

- **Results**

According to η_o values, the fiber orientation distribution in nonwovens explains the corresponding anisotropy in the composite properties. The Figure 5 and Figure 6 show the comparison between measured and predicted tensile moduli of composites in CD and MD directions. A good agreement between the experimental and theoretical values of tensile modulus is observed. The regression model tends to be closer to the RoHMs model. Therefore, the regression model could be assimilated as a hybrid model. The models used here take into account the fiber properties and the anisotropy of nonwoven fabrics. These parameters have an important influence on the tensile modulus of nonwoven reinforced composites.



Error Bars: ± Standard Deviation

Figure 5. Measured and predicted tensile modulus in CD direction.

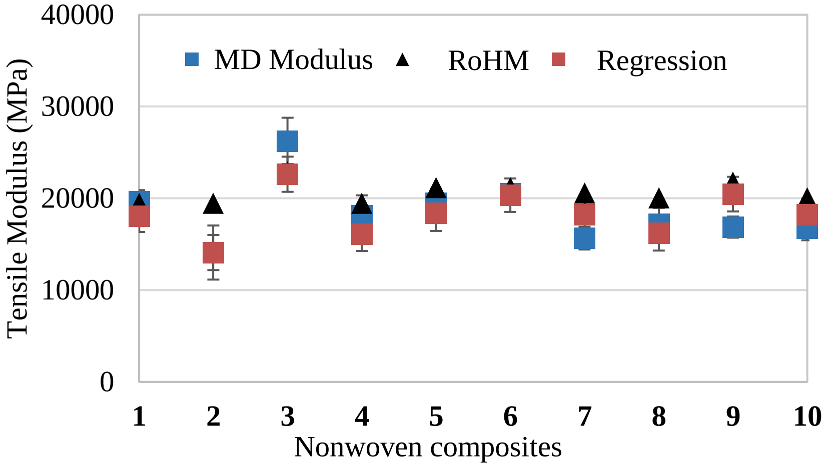


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Figure 6. Measured and predicted tensile modulus in MD direction**.**

4. Conclusions

In this study, the tensile properties of ten different carbon fiber nonwoven composites were investigated. The fiber type and length have great influence on the mechanical properties of the composites. The fiber orientation in nonwoven fabrics leads to the corresponding anisotropy in the composite properties. The tensile modulus and tensile strength of the composites produced are influenced by both tensile properties and architecture of fibers.

Theoretical analysis of the tensile modulus of nonwoven reinforced composites are discussed. The tensile moduli of the composites are predicted using regression model and RoHMs equation. Good agreements are observed between the experimental modulus and the theoretical results.

Further work is being undertaken to investigate the mechanical performance of recycled carbon fiber nonwoven reinforced composites.

Acknowledgments

The authors gratefully acknowledge the funding supports of “Direction Générale de l’Armement” and “Alpha Recyclage Composites”.

References

[1] O. Géraldine, L. Dandy and G. Leeke. Current status of recycling of fibre reinforced polymers: review of technologies, reuse and resulting properties. *Progress in materials science*, 72:61–99, 2015.

[2] P. Soraia and T.P. Silvestre. Recycling carbon fibre reinforced polymers for structural applications: Technology review and market outlook. *Waste Management*, 31: 378–392, 2011.

[3] M. Boulanghien, S. Da Silva, F. Berthet, G. Bernhart, Y. Soudais. Using steam thermolysis to recycle carbon fibres from composite waste. *JEC Compos Mag*, 2015; 100: 69–70.

[4] S.Y. Ye, A. Bounaceur, Y. Soudais and R. Barna, Parameter optimization of the steam thermolysis: a process to recover carbon fibers from polymer-matrix composites, *Waste Biomass Valor*, 4: 73-86, 2013.

[5] SJ Pickering, Z Liu, TA Turner and KH Wong, Applications for carbon fibre recovered from composites. *IOP Conference Series: Materials Science and Engineering*, 139:1, 2016.

[6] ELG Carbon Fibre Ltd, « http://www.elgcf.com/ ».

[7] J. Cornell. Experiments with Mixtures: Designs, Models, and the Analysis of Mixture Data. 2011.

[8] N. Venkateshwaran, A. Elayaperumal and G.K. Sathiya. Prediction of tensile properties of hybrid-natural fiber composites. *Composites Part B*, Vol 43: 793-796, 2012.

[9] M,R,Mansor. Stiffness Prediction of Hybrid Kenaf/Glass Fiber Reinforced Polypropylene Composites using Rule of Mixtures (ROM) and Rule of Hybrid Mixtures (RoHM). *Journal of Polymer Materials*. 30: 321-334, 2013.

[10] P. Zhang. Hybrid effect on mechanical properties of M40-T300 carbon fiber reinforced Bisphenol A Dicyanate ester composites. *Polymer Composites*, 31: 2129–2137, 2010.

[11] H.L. COX. The elasticity and strength of paper and other fibrous materials. Britis*h Journal of Applied Physics*, 3:72, 1952.

[12] H. Krenchel. Fibre Reinforcement: Theoretical and Practical Investigations of the Elasticity and Strength of Fibre-reinforced Materials. Copenhagen: Akademisk Forlag, 1964.

[13] ShHS. Yousfani, RH. Gong and I. Porat. Manufacturing of Fibre glass Nonwoven Webs Using a Paper Making Method and Study of Fibre Orientation in These Webs. *FIBRES & TEXTILES in Eastern Europe,* 20, 2(91): 61-67, 2012.