

Hybrid effects on effective mechanical properties of CF/FF and BF/FF epoxy-based composites

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Abstract. The paper explores the synergistic effects due to hybridization of different synthetic reinforcements with flax fibres on the effective mechanical properties of resulted compounds. A commercial epoxy polymer DGEBA is depolymerized as embedding material. Accounting for different stacking sequences, from symmetrical to unsymmetrical architectures, the samples were subjected to 3-point bending and their effective mechanical properties retrieved. Positive hybrid effects in the range of 9,5% and 35% for BF/FF and CF/FF architectures, were encountered in the hybrid composite specimens, respectively.

1. Introduction

Natural fibre-reinforced composites captured researchers' attention through-going over the last decade. Driven either by economic and social reasons or inherent yearning to state and bring an argument on their sustainability, these materials were intensively investigated both for materials' properties and behaviour with a relatively large area of engineering applications.

Literature survey provides evidence on various combinations between natural fibres and polymer matrices. Particular to the mechanical performance of these material types, the outstanding reviews of Shah (2013), Gurunathan (2015), Jauhari (2015) and more recently of Pickering (2016) and their co-authors can be used for further insights and issue development [1-4]. Readers get accustomed not only to a large spectrum of reinforcement and matrix materials' combinations and factors limiting their performance in action, but experimentally gathered data on mechanical properties from different testing configurations.

Moreover, arguments on natural materials' potential benefits, deployment of surface conditioning measures to improve the fibre/matrix adhesion, addition of chemical modifiers as cross-linking promoters and other procedures to limit the penalties of the resulting composite material performances were intensively tested and extensively debated [5-8]. Despite these, the issues of commercially available and practical use remain an intricate and yet to be implemented at production level desire.

In addition, since through hybridization improvements on the combination's effective properties were mostly achieved by individual material selection, both fibres and matrix, by smart reinforcement layering or intimately connecting, predictability about the preferences on the composite architectures adopted by different researchers' groups and lately by various industry players worldwide can be easily identified [9-12].

The green polymer-based composites developed hitherto were deploying natural fibres acquired from cellulose/ lignocellulose sources embedded mainly within affordable polymer resins. Scaling to



hybrid architectures augmented various combinations with synthetic reinforcements to address minimum requirements as structural elements or functional materials. The influence of constitutive, layering sequences, debates on fibre/ matrix interface issue upon the developed architectures' effective properties (e.g. mechanical, dynamical, thermo-physical, electrical, degradation, etc.) can be traced from already published reports [13-20].

The herein author has already approached some of the above issues on several hybrid composites out of glass and carbon fibres reinforcements and unsaturated polymer resin while investigating the influence of layering sequencing and fibre orientation upon their mechanical, dynamic and thermal effective material properties [21-22].

The present paper explores the feasibility of tailoring different hybrid architectures from natural- and synthetic-fiber reinforced polymer based prepreps, other than selected materials and tested combinations reported or delivered to the public at large. The synergetic effect revealed through hybridisation will be emphasised individually for each stacking sequence and material combination.

2. Experimental procedure

Basalt (n. BF), untreated flax (n. FF) and plain 1/1 weave carbon-fibre (n. CF) fabrics were used as reinforcements for the hybrid composite architectures. All reinforcements can be ranked as having a balanced distribution along the fabric's warp and weft directions. A commercial DGEBA epoxy resin (i.e. Epikote™ 04434) was cured with its delivered hardener under a ratio of 100 to 45 parts by weight of each constituent. The resin was chosen due to its wide availability and high thermoforming stability during laminate manufacturing. The hybrid composite laminates were produced by differently stacking nine pre-impregnated sheets out of natural and synthetic reinforcements.

Concerning the stacking sequence, materials exhibiting the highest strength values (i.e. CF, BF) were layered as external and external/ middle layers, giving rise to symmetrical and unsymmetrical hybrid architectures. Flax fibres were embedded in-between due to intrinsic poor material properties comparatively with the synthetic reinforcements. Table 1 lists the stacking layering codes used to further address the hybrid composite architectures and summarise their individual and total volume fraction within the final laminate.

The hybrid composite specimens were subjected to the flexural testing, according to DIN EN ISO 14125:1998 standard, using a mechanical testing machine INSTRON model 3369 running in a 3-point bending mode with a length span of 48 mm and 10 kN load cell. A bending rate of 1.5 mm/min was used to apply the load and an average out of five representative samples is provided. All mechanical tests were conducted at room temperature (approximately 21 ± 2 °C). Flexural strength and modulus were retrieved as claimed by the ISO 178:2010 standard.

The morphologies of composite architectures were examined deploying a scanning electron microscopy technique (SEM) on an EVO MA 25 (Zeiss, D) device, at room temperature, running in the 2.00 K x magnification mode (see figure 1 and figure 2 on representative specimens).

Table 1. Details on hybrid composites stacking sequences, assigned codes and volume fractions.

Stacking sequence	Laminate codes	Reinforcements volume fraction (vol %)		Total fibre loading (vol %)
		<i>nf</i>	<i>sf</i>	
□□□□□□□□	9FF	40	-	40
■□□□□□□■	BF/7FF/BF	21	14	35
■□□■□□■	BF/3FF/BF/3FF/BF	19	21	40
■□□□□□■	CF/7FF/CF	24	11	35
■□□■□□■	CF/3FF/CF/3FF/CF	23	12	35

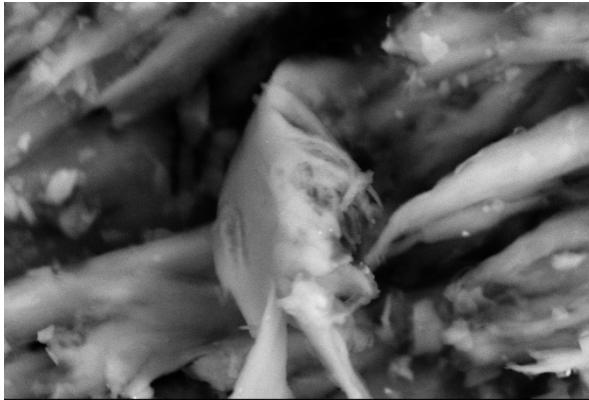


Figure 1. SEM micrograph on FF reinforced DGEBA laminate.

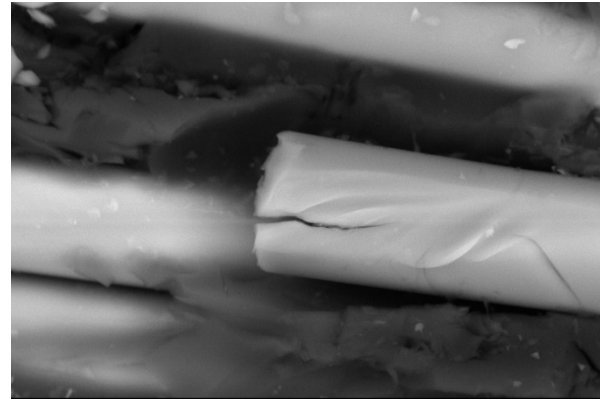


Figure 2. SEM micrograph on unsymmetrical BF/FF reinforced hybrid laminate.

3. Rule of mixtures (RoM) and hybrid rule of mixtures (RoHM)

Rules of mixtures (RoM) and hybrid mixtures (RoHM) were used to predict the effective stiffness of individual substrates and different stacked sequences of FF and CF and BF combinations, respectively. Expressions are functions of the composites' constitutive volume fraction and corresponding to individual material properties:

- RoM for natural-fibre reinforced composites

$$E_{nfc} = E_{nf}V_{nf} + E_m(1 - V_{nf}) \quad (1)$$

- RoM for synthetic-fibre reinforced composites

$$E_{sfc} = E_{sf}V_{sf} + E_m(1 - V_{sf}) \quad (2)$$

- RoHM for natural/synthetic hybrid composites

$$E_c = E_{nfc}V_{cnf} + E_{sfc}V_{scf} \quad (3)$$

In the above expressions, the indices stand for: *nf* – natural fibre, *sf* – synthetic fibre, *nfc* – natural fibres reinforced composite, *sfc* – synthetic fibres reinforced composite and *c* – hybrid composite, respectively.

Departure from the RoHM predictions of the experimental values from the symmetrical/unsymmetrical combinations contributes to the hybrid effect quantification, which in turn can be ranked as positive or negative [19]. This hybrid effect highlights the synergy reflected on the mechanical properties by natural and synthetic fibres combinations.

4. Results and discussions

Polymer composite behaviour in flexure can be regarded as depending both on the reinforcements and matrix material performance as well as on the fibre/matrix interface [14, 20]. The more optimum the fibre content, the more enhanced the stress transfer from polymer matrix to the fibres, and thus, the overall mechanical property aimed.

Prior insights in the mechanical behaviour of herein composite specimens, recalling the SEM images from figures 1 and 2, respectively, the enhanced adhesion of epoxy polymer to the BF against the fragility of FF fibres after the manufacturing process can be acknowledged.

As can be seen in figure 3, hybrid composite specimens, irrespective of CF or BF layers and stacking sequence, perform better in flexure compared with the FF reinforced composite samples. The load vs. elongation curves reveal increased slope with the increase of the synthetic reinforcement content and thus an increase of the effective elastic modulus and flexural strength.

Data collected in table 2 or represented in figure 4 reflect the above statements. Thus, the outperforming attributes of CF/FF hybrid architectures has to be assigned to the presence of CF layers. An increased content of CF layers results into both enhanced modulus and flexural strength.

On the other hand, an increasing tendency was expected on the percentage elongation at break values with the increase of synthetic reinforcement's content. The latter seems to reveal a negative effect and it has to be regarded to the FF and not to the CF or BF contents.

Table 2. Flexural properties of FF and hybrid composites.

Laminate codes	Flexural strength (MPa)	Strain at break (%)	Max. load (N)
9FF	169.55 ± 14.77	2.79 ± 0.27	226 ± 17.03
BF/7FF/BF	251.65 ± 7.61	4.07 ± 0.36	411 ± 19.98
BF/3FF/BF/3FF/BF	317.59 ± 23.63	3.74 ± 0.56	552 ± 36.46
CF/7FF/CF	350.95 ± 12.92	3.16 ± 0.65	467 ± 21.51
CF/3FF/CF/3FF/CF	358.15 ± 34.79	2.18 ± 0.10	488 ± 56.62

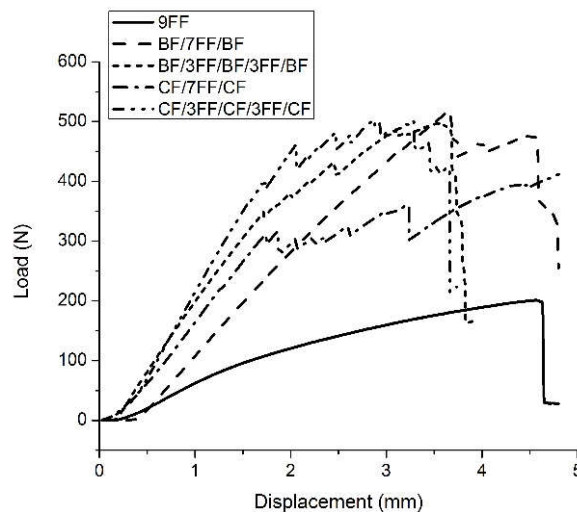


Figure 3. Load vs. displacement curves of composite specimens.

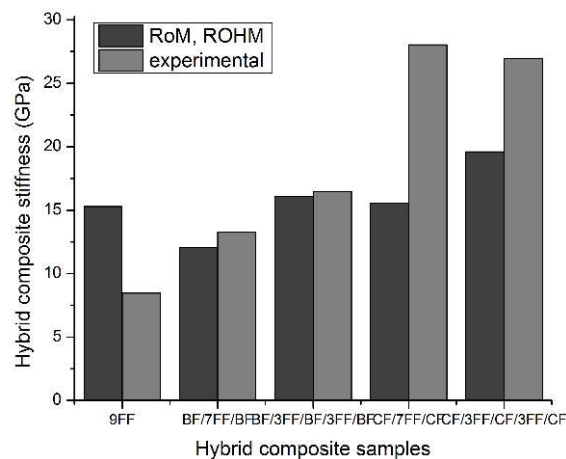


Figure 4. Theoretical vs. experimental stiffness values of samples.

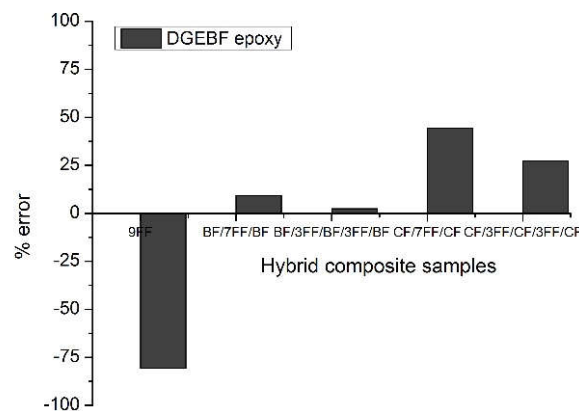


Figure 5. Percent error to aid hybrid effect characterisation.

Fibre/matrix interface has to be accounted further for these discrepancies in the mechanical properties. Thus, improper wetting and poor adhesion could be responsible for the above in addition to bending and shear failure mechanisms present with the outer and inner individual layer surfaces during the 3-point bending testing [17]. In this study, those above can be traced from accompanying SEM images with clear evidence on the inadequacy of FF fibres' outer surface.

Positive departures are obtained on BF and CF reinforced hybrid composites, either symmetrically or unsymmetrically stacked, in connection with flexural strength and elastic modulus, followed as performance indicators in the debate. Thus, the overall elastic moduli of BF hybrid architectures exhibited higher values compared with those from the FF laminate referential, with approximately 45% and 65%, respectively.

On the other hand, with CF/FF hybrid architectures, the synergetic effects are more pronounced in comparison with the BF/FF combinations, highlighting the efficiency of CF as reinforcement material. Little discrepancies were found between the symmetrical and unsymmetrical architectures, particularly the different individual and total volume fractions of reinforcements. Thus, property improvement increments by approximately 107% and 105%, for symmetrically and unsymmetrically stacked CF reinforced DGEBA composites over the FF laminate was identified, as it can be seen from data plotted in figure 4.

Further on the previous and concerning the hybrid effect, it can be accounted for from percent error values between experimentally retrieved and theoretically predicted values represented in figure 5. As it can be sized, all hybrid composite architectures reveal a positive hybrid effect on their performance indicator, with the same decreasing trend as the increase of the synthetic fibre content (e.g. from 9.3 % to 2.4 % for the BF/FF specimens). Once more, CF/FF hybrid architectures disclose their mechanical performance over the FF and BF/FF composites, owing to the CF reinforcement.

5. Conclusions

The paper aimed at debating on the effective mechanical properties and hybrid effects of several stacked sequences of synthetic (i.e. CF or BF) and natural (i.e. FF) fibre reinforced laminates based on a commercial DGEBA resin.

The synergetic effects, due to individual synthetic or natural reinforcements and various stacking sequences were tackled based on their flexural mechanical properties. The following conclusions have been drawn based on the predicted and retrieved data:

- improvements in the mechanical performances can be underlined for all hybrid composite architectures herein, irrespective of the constitutive stacking sequence and reinforcement material deployed;
- as expected, CF reinforced hybrids performed better in comparison with BF reinforced combinations for the same stacking sequence, whereas the latter appears to be an insurmountable competitor when cost issues come into focus;
- the results on overall material properties showed that symmetrically stacked CF reinforced architectures reveal similar values with unsymmetrical BF hybrid composites.

This research is expected to contribute effectively to CF and BF reinforced polymer composites market in search for novel routes for obtaining outstanding performance/cost ratios from their applications that can be sought in the area of constructions, automotive industry, transports, etc.

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References

- [1] Shah D 2013 *J. of Material Science* **48** 6083
- [2] Gurunathan T, Mohanty, S and Nayak S 2015 *Composites: Part A* **7** 1
- [3] Pickering K, Efendy M and Aruan L T 2016 *Composites Part A: Appl. Sci. Manuf.* **83** 98
- [4] Jauhari N, Mishra R and Thakur H 2015 *Materials Today: Proceedings* **2** 2868
- [5] Jawaid M and Khalil A H 2011 *Carbohydrate Polymers* **86(1)** 1
- [6] Swolfs Y, Gorbatiikh L and Verpoest I 2014 *Composites Part A: Appl. Sci. Manuf.* **67** 181
- [7] Jawaid M, Khalil A H and, Alattas O 2012 *Composites Part A: Appl. Sci. Manuf.* **43(2)** 288
- [8] Dicker M P M, Duckworth P F, Baker A B, Francois G, Hazzard M K and Weaver P M 2014 *Composites Part A: App. Sci. Manuf.* **56(0)** 280
- [9] Pil L, Bensadoun F, Pariset J and Verpoest I 2016 **83** 193
- [10] LeGault M 2016 *Composites World* 68
- [11] Cheung H, Ho M, Lau K, Cardona F and Hui D 2009 *Composites Part B: Engineering* **40(7)** 655
- [12] Dittenber D and Ganga Rao H 2012 *Composites Part A: App. Sci. Manuf.* **43(8)** 1419
- [13] Praveen R, Jacob S, Murthy C, Balachandran P and Rao Y 2011 *Cryogenics* **51(2)** 95
- [14] Muralidhar B 2013 *Mat. & Design* **52** 835
- [15] Zhu J, Njuguna J, Abhyankar H, Zhu H, Perreux D and Thiebaud F 2013 *Ind. Crops and Prod.* **50** 68
- [16] Lazko J, Landercy N, Laoutid F, Dangreau L, Huguet M and Talon O 2013 *Polymer Degradation and Stability* **98(5)** 1043
- [17] Assarar M, Zouari W, Sabhi H, Ayad R and Berthelot J 2015 *Composite Structures* **132** 148
- [18] Bertomeu D, García-Sanoguera D, Fenollar O, Boronat T and Balart R 2012 *Polymer Composites* **33(5)** 683
- [19] Marom G, Fischer S, Tuler F and Wagner H 1978 *J. of Mat. Sci.* **13(7)** 1419
- [20] Ozturk S 2005 *J. of Mat. Sci.* **40(17)** 4585
- [21] Motoc Luca D, Ferrandiz Bou S and Balart R 2015 *J. of Comp. Mat.* **49(10)** 1211
- [22] Motoc Luca D, Ferrandiz Bou S and Balart R 2018 Thermal properties comparison of hybrid CF/FF and BF/FF cyanate ester-based composites *J. of Thermal Analysis and Calorimetry* 10.1007/s10973-018-7222-y