Influence of the Manufacturing Process on the Tensile Stress-Strain Response of Hybrid Glass/Carbon and Carbon/Carbon Composites

Zahir Namourah, Filipe Ribeiro, and José Sena-Cruz(✉)

Department of Civil Engineering, ISISE/IB-S, University of Minho, Guimarães, Portugal
jsena@civil.uminho.pt

Abstract. Despite the numerous advantages of fibre reinforced polymers (FRP) composites, ductility is still a major problem of these materials. Usual FRP composites are stiff and strong with little or no warning before final failure. The mentioned drawback can be mitigated using unidirectional (UD) hybrid composites (i.e. composites in which two or more different reinforcing materials are combined in the same polymeric matrix). In these materials the development of tensile pseudo-ductile behaviour during the failure process can be achieved. The amount of resin used to manufacture hybrid FRP composites is responsible for significant changes at their tensile stress-strain curve. It is believed that these changes are dependent on the interlaminar fracture toughness of the interface between layers. In the present work, the effect manufacturing methods on the tensile properties of hybrid composites was studied. Hand lay-up and vacuum bagging techniques were compared. Three combinations of dry unidirectional fabric materials were used to produce hybrid FRP composites, namely: i) high-modulus carbon, ii) standard carbon, and iii) E-glass. An epoxy-based resin was used as matrix. Failure modes, tensile elastic modulus, strength, and stress-strain curve were analysed. Finally, experimental results were analytically simulated.

Keywords: Hybrid composite · Hand Lay-up · Vacuum bagging · Analytical analysis

1 Introduction

The tensile stress-strain response of UD hybrid composites is one of the most interesting characteristics of these materials. In fact, it was found that hybridisation can promote a ‘pseudo-ductile’ tensile response. This behaviour is characterized by fragmentation of low strain (LS) material and dispersed delamination of the LS material fragments from the undamaged high strain (HS) material.

During the last years, the authors of the present work have been studying the use of UD interlayer (layer-by-layer) hybrid composites in civil engineering (Namourah 2019; Ribeiro 2019; Ribeiro et al. 2018). Despite the work carried out, the influence of the manufacturing method of hybrid composites on their tensile mechanical properties still a remaining open question. In the present work, the influence of manufacturing method,

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namely hand lay-up and vacuum bagging, on the tensile behaviour of UD interlayer glass/high modulus carbon and standard-modulus carbon/high modulus carbon hybrid FRP composites was studied. Hand lay-up method is a manual method for producing composites, requiring simple tools. Despite of its simplicity, it lacks the ability of controlling the required amount of resin and avoiding defects and voids. Vacuum bagging is an improved impregnation method since it increases fibre content of the laminates and it lowers the void contents. Additionally, this method is healthier and safer since it reduces the amount of volatiles emitted during cure process.

The obtained results were critically analysed. Finally, experimental results were analytically simulated using based on existing models.

2 Experimental Work for Assessing Hybrid Composites

2.1 Experimental Programme

Three types of commercial dry fibre fabrics were used, namely: i) unidirectional high modulus carbon (S&P C-Sheet 640), ii) unidirectional standard carbon (S&P C-Sheet 240), and iii) bidirectional (90% in longitudinal and 10% in transversal direction) E-Glass (S&P G-sheet E 90/10).

An epoxy-based material, S&P Resin 55 HP, was used as matrix for laminating the studied composites, as recommend by the supplier of three dry fabrics.

Table 1 includes the density, areal mass, fibre layer thickness (areal mass density divided by the volumetric mass density) and the basic tensile properties (elastic modulus, tensile strength and strain at the failure) of the mentioned materials assessed by the manufacturer.

Table 1. Properties of the dry fibre fabrics.

Fabric ID	Density [g/m^3]	Areal mass [g/m^2]	Layerthickness [mm/layer]	Elastic modulus [MPa]	Tensile strength [MPa]	Strain at the failure [%]
HM carbon (HMC)	2.10	430	0.205	$\geq$ 640000	$\geq$ 2650	0.40
ST carbon (C)	1.79	430	0.240	$\geq$ 240000	$\geq$ 3400	1.70
E-glass (G)	2.60	440	0.169	$\geq$ 73000	$\geq$ 3400	4.50

Three different combinations were adopted being the middle layer of fibre fabric always the high modulus carbon fabric, namely: i) G/HMC/G; ii) G/G/HMC/G/G; and, iii) C/HMC/C. The selection of the three combinations was based on the results obtained by Ribeiro et al. (2018), since among all the combinations tested by the authors, this maximized the pseudo-ductile behaviour. For each manufacturing process (hand lay-up and vacuum bagging), two replications were produced. Which series was composed of four specimens.

The series were dominated according to the following criteria (Table 2): i) adding the letters of H or V to indicate the manufacturing process, hand lay-up or vacuum bagging, respectively; ii) adding the number 1 or 2, representing the number of series replication; finally, iii) adding a number to identify the specimen in the same series (from 1 to 4). A total of 48 specimens (3 material combinations × 2 manufacturing processes × 2 replications × 4 specimens) were tested.

Table 2. Sample domination of the tested hybrid composites.

Series ID [domination]	Stacking sequence	Method [domination]	Series [domination]	Sample			
				1	2	3	4
G/HMC/G [1G]	■□■	Hand lay-up [H]	Replication 1 [1GH1]	1GH11	1GH12	1GH13	1GH14
			Replication 2 [1GH2]	1GH21	1GH22	1GH23	1GH24
		Vacuum bagging [V]	Replication 1 [1GV1]	1GV11	1GV12	1GV13	1GV14
			Replication 2 [1GV2]	1GV21 1GV21	1GV22	1GV23	1GV24
G/G/HMC/G/G [2G]	■■□■■	Hand lay-up [H]	Replication 1 [2GH1]	2GH11	2GH12	2GH13	2GH14
			Replication 2 [2GH2]	2GH21	2GH22	2GH23	2GH24
		Vacuum bagging [V]	Replication 1 [2GV1]	2GV11	2GV12	2GV13	2GV14
			Replication 2 [2GV2]	2GV21	2GV22	2GV23	2GV24
C/HMC/C [C]	■□■	Hand lay-up [H]	Replication 1 [CH1]	CH11	CH12	CH13	CH14
			Replication 2 [CH2]	CH21	CH22	CH23	CH24
		Vacuum bagging [V]	Replication 1 [CV1]	CV11	CV12	CV13	CV14
			Replication 2 [CV2]	CV21	CV22	CV23	CV24

Notes: HS – high modulus; LS – low modulus; ■– HS fibres layer; □– LS fibres layer

2.2 Hand Lay-Up Method

The hybrid composite laminates manufactured by hand lay-up method followed the main steps suggested by the guidelines, e.g. CNR-DT200 (2013). All the specimens were produced in a rigid plate of glass cover with a Teflon film. As example, the steps are given for series '2G':

a) Cutting fabrics of 300 mm by 150 mm;
b) Preparing the resin according to the manufacturer's technical data sheet;

c) Laying a layer of the resin on Teflon film;
d) Impregnation of the first E-glass fabric layer and then lay-up it on the Teflon film;
e) Impregnation of the second E-glass fabric layer and then lay-up it on first E-glass fabric layer;
f) Impregnation of the high modulus carbon fabric and lay-up it on the first two glass fabrics;
g) Repeat the similar process of e) for the fourth and fifth E-glass fabric layers;
h) Finally, rolling and applying uniform pressure on the all layers of the composite using a rolling tool.

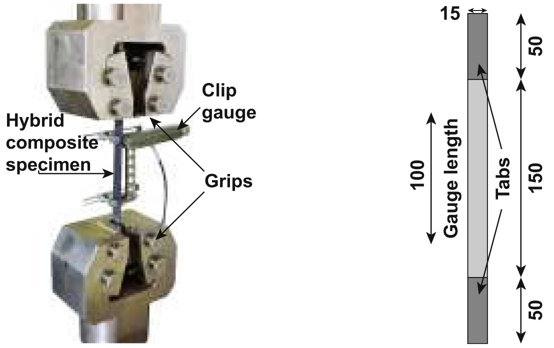
2.3 Vacuum Bagging Method

Like in the previous case, the production of the hybrid FRP composite laminates by vacuum bagging follows the best practices, e.g. (“Vacuum Bagging Equipment” 2019). Thus, the following protocol was followed in the manufacturing of hybrid composites:

a) Steps a) to g) described in the ‘Hand lay-up Method’;
b) Apply the first separation permeable layer on the impregnated composites;
c) Applying of the cotton layer;
d) Applying the second separation permeable layer;
e) Cover all the layers with plastic;
f) Ensure the complete closure and adjustment of the covered plastic with the glass plate;
g) Insertion of the vacuum bagging pipe with complete closer assurance;
h) Applying the vacuum.

2.4 Test Protocol

Tensile tests were performed according to ISO 527–5 (2009), at room temperature, on a universal testing machine with a 200 kN load cell (linear error less than 0.05% of full scale) and hydraulic grips, as shown in Fig. 1. The specimens have a total length of 250 mm and width of 15 mm. Prior the execution of the tests, aluminium tabs (50 mm × 15 mm × 1.5 mm) were glued at the extremities and both faces to prevent gripping effects. A clip gauge (linear error of 0.25%) with an initial gauge length of 100 mm was placed at the central part of the specimens. The tensile tests were performed at a rate of 1 mm/min up to failure.



a) b)

Fig. 1. Tensile test: a) illustration of the test, b) geometry of composite specimen. (Dimensions in [mm]) (Ribeiro et al. 2018).

3 Results and Discussion

Typical failure modes of the tested composites are depicted in Figs. 2 and 3, while stress-strain diagrams of the three tested combination are presented in Fig. 4. Elastic modulus (E-modulus) and tensile strength are included in Table 3. The calculations of the involved parameters followed ISO 527–1 (1993): i) tensile stress (σ) as the ratio between the tensile force and the area of the original cross-section within the gauge length; ii) tensile strain (ε) is the increase in length per unit original length of the gauge; iii) E-modulus (Eexp.) was defined as the slope of the line defined by two points of the stress-strain curve (strain $\varepsilon 1 = 0.0005$ and strain $\varepsilon 2 = 0.0025$); iv) tensile strength (σexp) as the tensile stress for the maximum tensile force.

Different failure modes between the two production methods were clearly observed. Failure modes of composites produced using hand lay-up method were characterized by multiple cracks of low strain material, i.e., carbon-layer, followed by localized delamination, until the final failure which represented by the arrow 9 (Fig. 2). Other arrows and their numbers in the figure represent the location and the consequence of each crack, respectively. On the other hand, composites produced using vacuum bagging method have failed by progressive delamination until the final failure that represented by the last two arrows (Fig. 3), after the formation of the first crack. Other arrows in the figure represent the location of the fed delamination.

According to the experimental results, it is clear that all composites produced by hand lay-up method have pseudo-ductile behaviour due to low strain material fragmentation and delamination, except the samples 1GH11, 1GH13, 1GH21, and 1GH22. In the case of composites produced by vacuum bagging method, the fragmentation of low strain material was not observed (Fig. 4).

From Table 3 it is possible to observe an increase in terms of the E-modulus and stress of the series produced with the vacuum bagging method, when compared with the hand lay-up one. This increase was expected given the same amount of dry fabrics and reduced amount of resin in the case of vacuum bagging method.

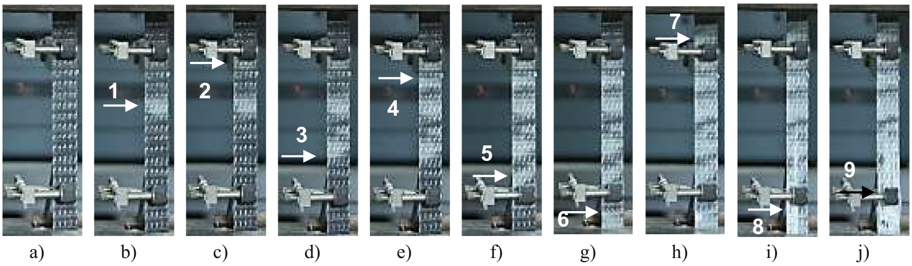


Fig. 2. Failure sequence of a sample specimen (2GH11) of G/G/HMC/G/G composite, produced by hand lay-up method.

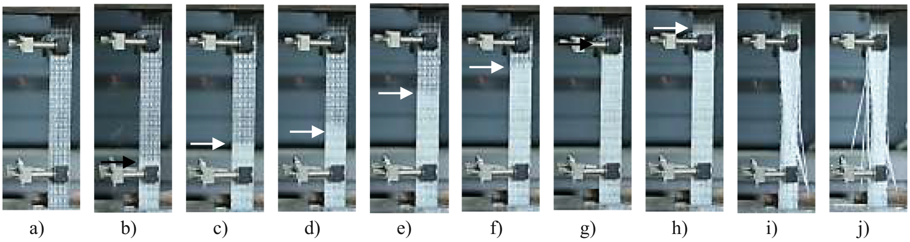


Fig. 3. Failure sequence of a sample specimen (2GV21) of G/G/HMC/G/G composite, produced by vacuum bagging method.

4 Analytical Modelling

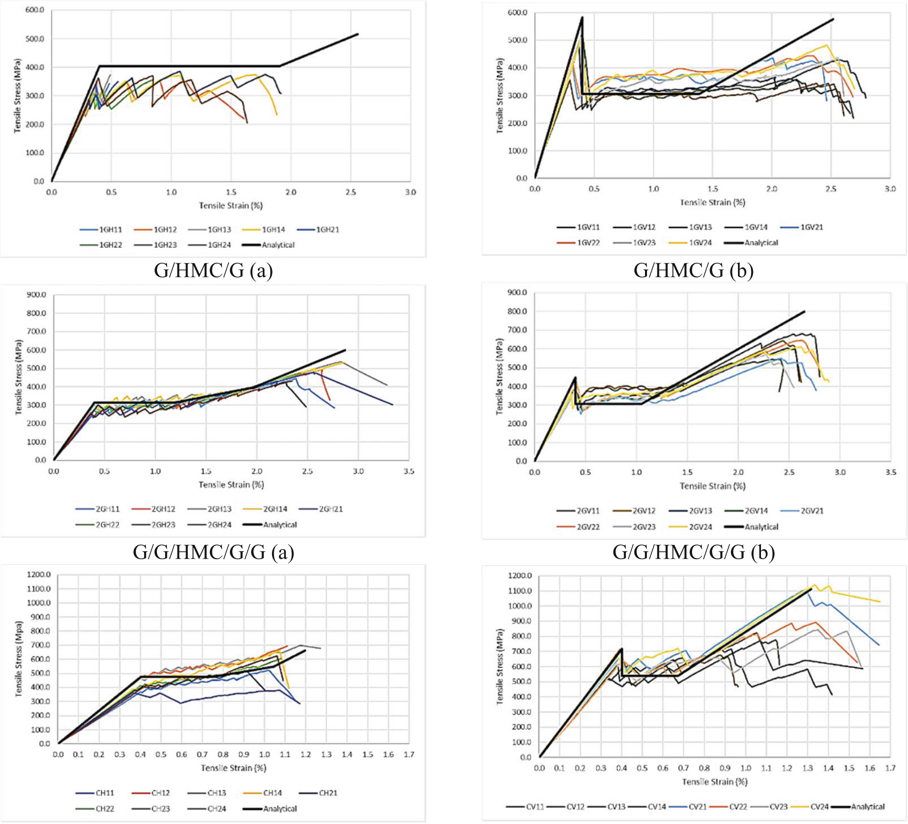
According to Jalalvand et al. (2015), Ribeiro et al. (2018), and You et al. (2017), the prediction of tensile properties of the hybrid composites can be achieved using the geometrical and mechanical properties of their main components.

Among the analytical models that can be used to predict the *E*-modulus, the rule of mixture model is one of most appropriated ones. However, for tensile strength prediction the bilinear rule of mixture method has been proposed. Moreover, accurate prediction of the overall behaviour of the hybrid FRP composite was proposed by Jalalvand et al. (2015). In this model, in addition to the *E*-modulus and the tensile strength, the whole behaviour of the hybrid FRP composite can be predicted from the first moment of loading up to failure in the stress-strain diagram. In the present work, this approach was followed.

4.1 Rule of Mixtures (ROM)

The prediction of the E-modulus of hybrid FRP composite can be obtained using the rule of mixtures (ROM) (Swolfs et al. 2014). ROM is the sum of the contributions of E-modulus of each raw material based on their volume fraction, according the following assumptions (You et al. 2017):

i. The fibres are homogenous, linear elastic, and well-arranged regularly in space;
ii. The matrix is homogenous, linear elastic, and isotropic;



G/HMC/G (a)

G/HMC/G (b)

G/G/HMC/G/G (a)

G/G/HMC/G/G (b)

C/HMC/C (a)

C/HMC/C (b)

Fig. 4. Experimental and analytical stress-strain diagrams, (a) using hand lay-up production method, (b) using vacuum bagging production method.

iii. The plies are microscopically homogenous, linear elastic, orthotropic; and, initially in a stress-free state;

iv. There are no voids; the fibre and the matrix are completely coupled.

Therefore, the *E*-modulus modulus of hybrid composite (E_{hybrid}) can be calculated according to the Eq. (1) (Swolfs *et al.* 2014):

$$E_{hybrid} = V_L E_L + V_H E_H + V_M E_M \, [MPa] \tag{1}$$

where V_L, V_H, V_M, E_L, E_H, and E_M are the volumetric fraction and E-modulus of the low strain fibres, high strain fibres and matrix, respectively.

4.2 Bilinear Rule of Mixtures

According to several studies (Kretsis 1987; Manders and Bader 1981; Pan and Postle 1996; Shan and Liao 2002; Swolfs et al. 2014), ROM can't be applied to predict tensile

Table 3. Experimental and analytical results.

Composite	Method	E_{exp}	E_{hybrid}	$E_{\text{Relative Error}}$	σ_{exp}	σ_{hybrid}	$\sigma_{\text{Relative error}}$
		[MPa] (CoV%)	[MPa]	[%]	[MPa] (CoV%)	[MPa]	[%]
G/HMC/G	Hand Lay-up	88052 (3.15)	97472	−10.7	315.0 (12.37)	392.6	−24.64
	Vacuum Bagging	127642 (4.98)	140971	−10.4	470.0 (13.07)	573.8	−22.08
G/G/HMC/G/G	Hand Lay-up	68974 (4.43)	75645	−9.7	282.9 (7.90)	301.9	−6.72
	Vacuum Bagging	98653 (5.96)	108210	−9.7	396.8 (6.03)	437.6	−10.28
C/HMC/C	Hand Lay-up	103120 (8.49)	114520	−11.1	406.1 (11.12)	457.7	−12.70
	Vacuum Bagging	165689 (4.97)	172855	−4.3	620.2 (10.53)	697.5	−12.47

strength. In reality, the spread of the fibre can be non-homogenous and the fibre orientation can be misaligned, resulting in reduced tensile strength of the unidirectional fibre composite (You et al. 2017). Moreover, since the fibre, with relatively high stiffness, and the matrix, with relatively low stiffness, exhibit different tensile behaviour, shear-lag may occurs and causes rupture to occur at different points of time. For this reason, ROM predicts the E-modulus of FRP with relatively good accuracy but overestimates the tensile strength (You et al. 2017).

To predict the tensile strength of hybrid composites, σ_{hybrid}, some authors have proposed a bilinear ROM, as shown in Eq. (2) (Aveston and Sillwood 1976; Kretsis 1987; Manders and Bader 1981):

$$\sigma_{hybrid} = \begin{cases} V_L S_L + V_H E_H \varepsilon_L;\ V_H < V_{crit} \\ V_H S_H;\ V_H > V_{crit} \end{cases} \text{[MPa]} \tag{2}$$

where S_L and S_H are the reference strengths of the low strain and high strain fabric fibres and ε_L is the strain at the failure of low strain fabric fibres. V_{crit}, as shown in Eq. 3, was calculated by equating the two branches of Eq. (2) taking into account that $V_L + V_H = 1$, i.e., VL is equal to 1-VH:

$$V_{crit} = \frac{S_L}{S_L + S_H - E_H \varepsilon_L} \quad [\%]. \tag{3.}$$

Table 3 shows the predicted values of the E-modulus (Ehybrid) according to the rule of mixture (ROM) previously described. In the same table, it is also presented the relative error between analytical predictions and the experiments. In general, good predictions can be observed with relatively low errors.

The table also shows the analytical predictions of the tensile strength using the bilinear ROM, σ_{hybrid}. These predictions are compared with experimental values, σ_{exp}, thought the relative error. All the necessary input parameters were provided by the manufacturer. In general, good predictions can be also observed with low relative errors, with the exception of series G/HMC/G where relatively high errors were obtained.

4.3 Stress-Strain Response of Hybrid Composites – Model of Jalalvand et al. (2015)

Previous methods are simple and they are capable of predict the E-modulus and tensile strength accurately but not the whole tensile stress-strain curve. Jalalvand et al. (2015) has proposed an analytical model to predict the stress–strain curve of hybrid composites. According to the proposed model, after the first crack of the low strain material, four different scenarios can occur:

i. Premature failure of the high strain material;
ii. Catastrophic delamination followed by high strain material failure;
iii. Fragmentation (multiple fractures) in the low strain material and then high strain material failure;
iv. Fragmentation in the low strain material followed by dispersed delamination and then high strain material failure.

The analytical modelling approach proposed by Jalalvand et al. (2015) takes into account these four damage modes. In his work, the estimation of three stress levels were proposed, namely i) the stress level at the fragmentation in the low strain material, $\sigma_{@LF}$ (Eq. (4)) and ii) the stress level at delamination, $\sigma_{@del}$, (Eq. (5)) and iii) the stress level at the failure of high strain material, $\sigma_{@HF}$ (Eq. (6)). These equations were upgraded in the scope of the present work.

$$\sigma_{@LF} = \frac{S_L}{E_L}\frac{E_L t_L + E_H t_H + E_M t_M}{t_L + t_H + t_M} = \frac{S_L E_{int}}{E_L} \quad \text{[MPa]} \tag{4}$$

$$\sigma_{@del} = \frac{1}{t_L + t_H + t_M}\sqrt{2G_{IIc}E_H t_H\left(1 + \frac{E_H t_H}{E_L t_L + E_M t_M}\right)} \quad \text{[MPa]} \tag{5}$$

$$\sigma_{@HF} = \frac{S_H}{K_t}\frac{t_H}{\sqrt[m]{V}(t_L + t_H + t_M)} \quad \text{[MPa]} \tag{6}$$

In these equations t_L, t_H, t_M; E_L, E_H, E_M; and S_L and S_H, are the half thickness; the E-modulus; and the tensile strength of the low strain; high strain; and matrix material, respectively. E_{int}, G_{IIc}, K_t, m, and V, are the E-modulus; the energy release rate; the stress concentration factor; Weibull modulus of high strain material strength distribution; and the whole high strain material volume, respectively.

Table 4 shows the input parameters of the model. Half thickness of matrix, t_M; is the half thickness value of subtracting fiber fabrics thinness from the total thickness. Length and width required for the volume of the high strain fibre, V, represents the measured mean length and width of the specimen. Energy release rate, G_{IIc}; stress concentration factor, K_t; and Weibull modulus of high strain material strength distribution, m; were selected according to the experimental behaviour of each composite (Jalalvand et al. 2015). Other geometrical and properties were as provided by the manufacturer.

Based on the characteristics points and the equations introduced in the previous section, it was possible to predict the stress-strain curves of the hybrid FRP composites. Figure 4 includes the predicted stress-strain curves of the hybrid FRP composites of

Table 4. Analytical stress levels of hybrid composites and their input parameters.

Composite	Method	t_M	E_M	S_M	G_{IIc}	K_t	m	V	E_{int}	$\sigma_{@LF}$	$\sigma_{@del}$	$\sigma_{@HF}$
		[mm]	[MPa]	[MPa]	[N/mm]	[-]	[-]	[mm3]	[MPa]	[MPa]	[MPa]	[MPa]
G/HMC/G	**Hand Lay-up**	0.545	3200	55.4		1.07	29.3	1267.64	97472	403.6		515.8
	Vacuum Bagging	0.287	3200	55.4	1.00	1.40	29.3	1278.08	140971	583.7	306.2	576.0
G/G/HMC/G/G	**Hand Lay-up**	0.785	3200	55.4	3.50	1.20	29.3	2558.20	75645	313.2	396.0	598.4
	Vacuum Bagging	0.405	3200	55.4	1.00	1.30	29.3	2557.88	108210	448.1	307.6	800.7
C/HMC/C	**Hand Lay-up**	0.754	3200	55.4	1.70	1.10	29.3	1791.27	114520	474.2	548.9	663.1
	Vacuum Bagging	0.377	3200	55.4	0.70	1.00	29.3	1800.37	172855	715.7	539.0	1111.5

all the series tested. The figure also includes the experiments. In general, there is a good agreement between predictions and experiments. Analytical estimations show that, before the last failure, i.e. the failure of high strain material, the three damage modes were obtained: catastrophic delamination, fragmentation, and fragmentation followed by delamination. All composites produced by vacuum bagging method, analysed presented catastrophic delamination, and then high strain material failure. Composites produced by hand lay-up method have damage mode of low strain material fragmentation and disturbed delamination, followed by high strain material failure, except in half of both replications' specimens of G/HMC/G composite, where they have premature failure of high strain material.

5 Conclusions

In the scope of the present work, the effect of the hand lay-up and vacuum bagging manufacturing methods on the tensile response of hybrid composites has been investigated. Three distinct hybrid composite configurations were tested. Several raw materials were used: high-modulus carbon, standard carbon and E-Glass were used and fibres and epoxy resin as matrix.

Three models were used to analytically predict the mechanical properties of hybrid FRP composites namely, *E*-modulus, tensile strength, and the whole tensile stress-strain curve of the composites.

From this study the following main conclusions can be drawn:

1. All composites produced by hand-layup method provide pseudo-ductile behaviour except ones that failed prematurely in the low relative volume of high strain material in 1GH combination;
2. Specimens produced by vacuum bagging yield to the premature delamination;
3. Enhanced mechanical tensile stress-strain properties were observed in the case of the vacuum bagging method, due to the reduction of resin content;
4. The low strain material always governed the first failure.

5. In general, the proposed analytical models have predicted mechanical response of all the composites with high accuracy.

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