

IMPROVEMENT OF FLAX FIBRES REINFORCED COMPOSITE BY HYBRIDIZATION WITH CARBON FIBRE: SORPTION AND MECHANICAL PROPERTIES

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Abstract

The purpose of this work is the improvement of flax fibres reinforced composites obtained by vacuum moulding. The relatively high degree of porosity in this kind of composites, due to the lack of compatibility between epoxy matrix and flax fibres, remains a major constraint for their use in the industrial world. Hence, we have used a combination of carbon and flax fibres in order to optimize the properties of the assembly. Several stacking sequences have been tested to analyse the influence of the addition of carbon fibres and basically their position inside the structure on the water recovery behaviour and the buckling test response.

1. Introduction

In the last decade, the use of bio-fibres to replace synthetic carbon/glass fibres as reinforcement in polymer composites has grown explosively and has found his way in several sectors such as technical, building construction and generally, engineering applications. Because of his environmental friendly culture and industry, low densities (lighter than carbon and nearly twice as light as glass fibres) and damping properties, flax fibres are a serious candidate and alternative to reinforce material composites. However flax fibre possess some limitations which are basically their highly hydrophilic behavior, their strong mechanical and physical property dependence on the location and the weather of the crop field, their bad adhesion with polymeric matrix and their thermal degradation above 200 °C. Much effort has gone into increasing their mechanical performance to extend the capabilities and applications of this material. Several previous works were performed as studying the effect of conventional textile treatments of flax fabric on the mechanical properties and the water sorption of flax/epoxy composites [1-4]. The bad adhesion of flax fibres leads to poor interfacial bonding limiting mechanical performance as well as low moisture resistance affecting long term properties. The other main problem faced with flax fibre reinforced composites is the highly porosity rate which affects considerably the mechanical properties of composites [5, [6]. The purpose of the presented research work is to investigate the effect of the hybridization of NFRC, by incorporation of carbon fibre fabrics into the stacking sequence, on the final composite behavior: water recovery and buckling test response. The concept of hybrid composite has been remarkably reviewed by Swolfs [7], and Sanjay compiled a fairly exhaustive review focused on hybridization of natural and artificial fibres, especially glass fibres [8]. Hybrid composite is defined as a matrix containing at least two different types of reinforcing fibres. Hybridization can be made at yarn or strand level by mixing fibres of different origins, at layer level with ply made of different yarns or strands, at stacking sequence level by alternating different plies. Assarar had used the concept

of hybridization of carbon and flax hybrid fibres to obtain composite material involving structural rigidity and vibration damping [9]. Bagheri had shown that incorporation of flax plies improves the fatigue strength of carbon composite used for bone fracture plate applications [10]. Dhakal had studied the effects of external carbon plies on flax composites [11]. He demonstrated that external carbon plies have a beneficial effect on water sorption and mechanical properties. It appears that hybridization of natural fibre layer with synthetic fibre layer is an effective and powerful method to combine their respective properties. Apart from the mechanical reinforcement, hybridization with synthetic fibres could be efficient to limit the sensibility of NFRC to moisture and environmental stresses. In the presented work, composites with various stacking sequences made of flax and carbon fibre fabrics were manufactured. The influence of carbon plies position on the hybrid composite performance had been studied with focus on the microstructures, the water sorption, and buckling test response.

2. Studied materials and processing methods

2.1. Flax fibre fabric and carbon fibre fabric

The flax cloth used is marketed by Flax technique under the trade name Twinflax 2D 235. It is a flax fibre woven fabric, with a basis weight of 235 g/m², using non-coated twisted strands while the carbon cloth is a 2/2 twill, with a 200g/ m² basis weight.

2.2. Epoxy matrix

The thermosetting polymer matrix is a mixture of an epoxy resin with a hardener. The epoxy resin, marketed under the name of Araldite LY 1564, is developed by the company Huntsman especially for impregnated structures and has a viscosity between 10 to 20 mPa.s at the temperature of 25 °C and a density of 1.1 to 1.2 g / cm³. The amine hardener developed by the same company under the name Aradur 3487 has a viscosity between 30 to 70 mPa.s at 25 °C and a density of 0.98 to 1 g / cm³. The optimal mix proposed by the manufacturer is 100 parts by weight of Araldite LY 1564 to 34 parts by weight of Aradur 3487 hardener.

2.3. Composite manufacturing

A composite plate is made of a total of eight plies of tissue. Plates of 8 flax plies, called 100% flax, were first carried out and served as a reference to determine the intrinsic properties of a laminate ply. Different stacking sequences have been tested mixing 4 plies of flax and 4 plies of carbon, all stacking sequences comply by mirror symmetry ([C C F F]_s, [C F C F]_s, [F C F C]_s, [C F F C]_s, etc). Some sequences were repeated several times.

The process used to manufacture the composite plates is the manual impregnation of plies followed by pressurization by the technique of the vacuum bag. After pressurization under 0.9 bar during 24 hours at room temperature, post-curing in an oven is required to obtain a complete polymerization of the epoxy matrix, which consists of two bearings; the first at 80 °C and the second at 130 °C for allows a complete cross-linking (Figure 1).

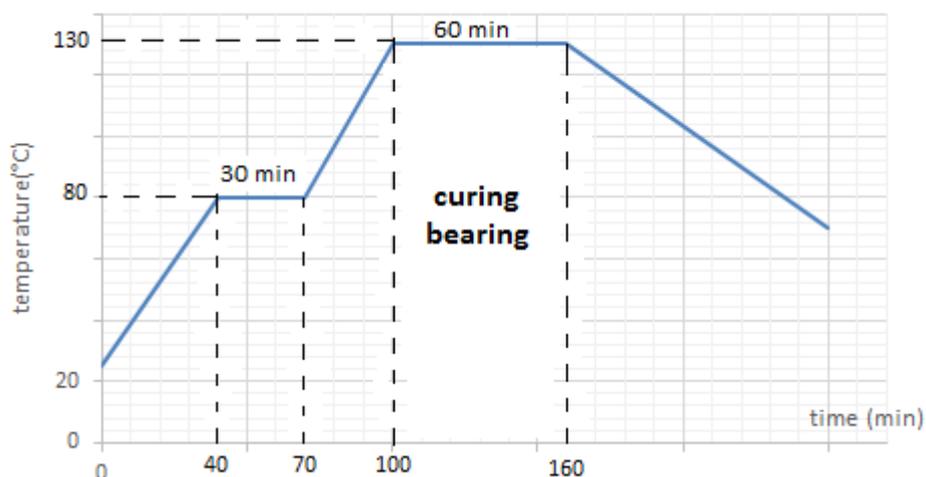


Figure 1. Post-curing cycle

3. Experimental methods

3.1. Sorption Test

Following ASTM D5229 norm, 120 mm square samples are cut in the composite plates. They are dried in an oven at 103 °C during 24 h to remove any moisture. Then, the sample four edges are covered by a thick layer of silicone paste, in order to prevent lateral edge sorption. It has been verified that silicone paste sorption is negligible compared to composite sorption. Then, the plates are immersed into a water bath at a controlled temperature of 20 °C and weighed regularly using an electronic weigh scale, 0.1 g of precision, in order to determine water content and then the sorption mechanism and the characteristic parameters such as saturation water content and diffusion coefficient. The water sorption rate M_t at time t can be expressed as follow:

$$M_t = \frac{W_t - W_0}{W_0} \quad (1)$$

where W_0 is the weight of dried specimen and W_t is the weight of the wet specimen at time t .

3.2. Buckling test

The buckling test is to determine the critical buckling load as well as the damage process of the several stacking sequences. The test stand is used an servo-hydraulic Instron 8800 tensile machine equipped with a 5kN force cell and mechanical self-tightening jaws. The tests are carried out at room temperature into quasi-static state with a travel speed of 2 mm/minute. The specimens are cut in the direction 0° and 45° of each plate using a diamond saw cooled with water. In absence of a standard for buckling test, was used to test standard NF T51-120-6 buckling fatigue.

4. Results & discussion

4.1. Water absorption

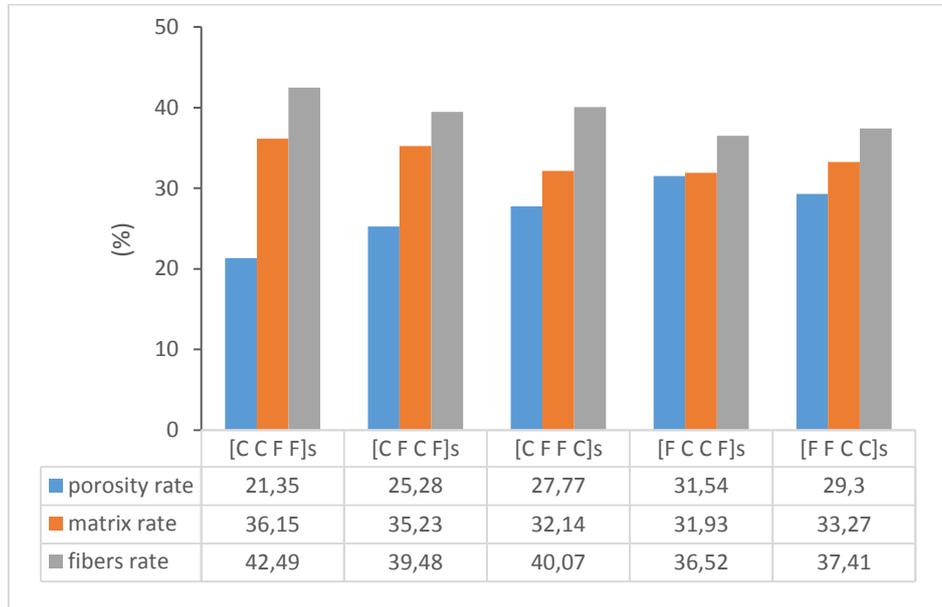


Figure 2. Volume rate of each component (matrix, fibre and porosity) of the different composite plates

Table 1. Characteristics of the sorption test

Plate stacking sequence	Fickian mechanism constant n	Porosity rate (%)	diffusion coefficient D ($\text{mm}^2.\text{s}^{-1}$)	Water content at saturation (%)
100% flax	0.453	32.4	4.72	32.9
[F C F C] _s	0.232	27.5	4.05	21.6
[C F C F] _s	0.340	26.34	2.11	20.09
[C C F F] _s	0.5	22.52	1.8	20

The experimental data were used to draw the water content M_t curve as a function of the root of time. These data are compared with the theoretical expression of Fickian diffusion for a large plate. The water absorption over time is given by equation (2) for the short time and the equation (3) for the long time [12].

$$\frac{M_t}{M_\infty} = \frac{4}{h} \sqrt{\frac{D \cdot t}{\pi}} \quad (2)$$

$$\frac{M_t}{M_\infty} = 1 - \frac{8}{\pi^2} \exp\left(\frac{-D \cdot \pi^2 \cdot t}{h^2}\right) \quad (3)$$

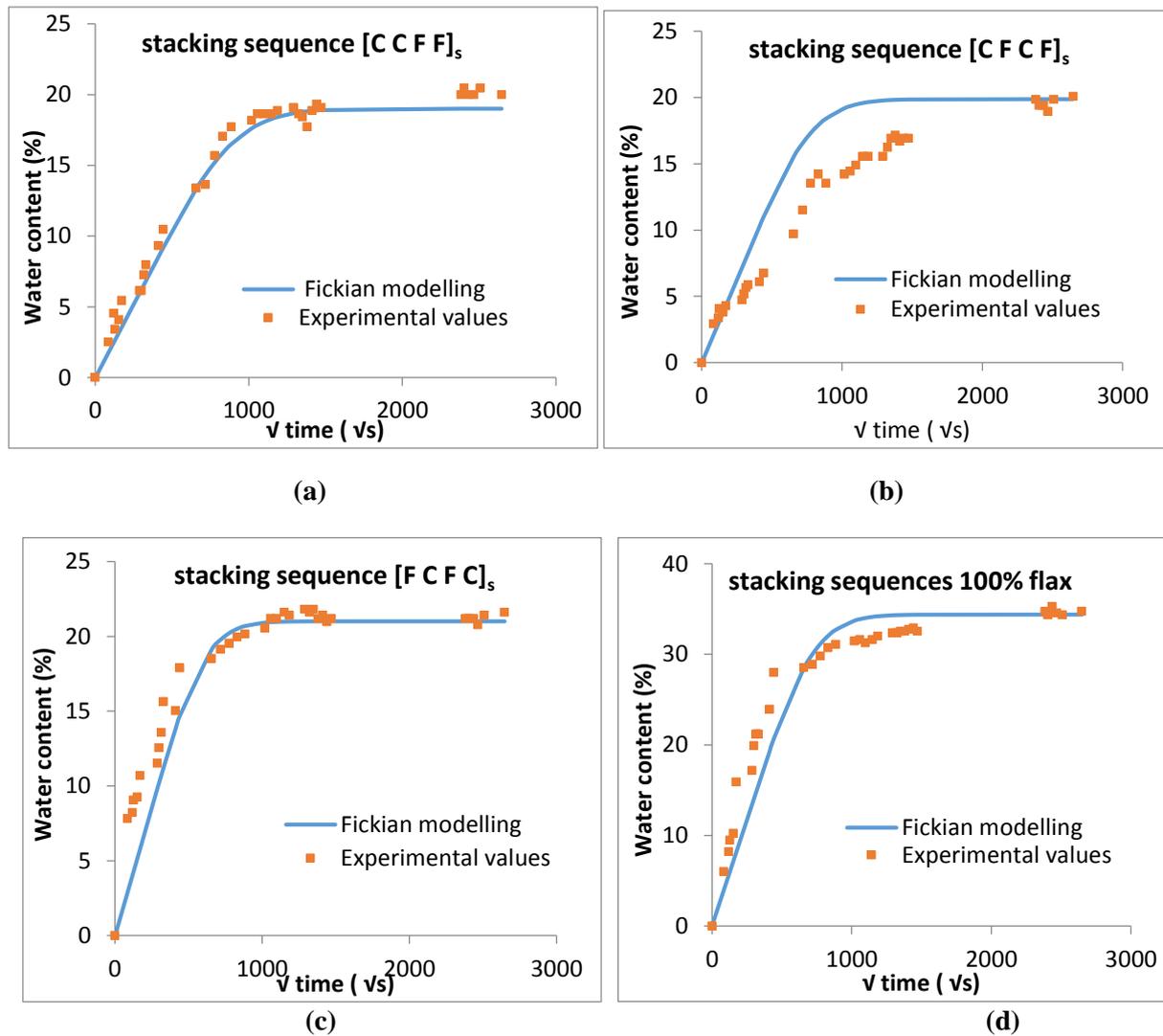


Figure 3. The classical Fick adjustment model on the experimental results of water sorption by the laminates (a) [C C F F]_s, (b) [C F C F]_s, (c) [F C F C]_s, (d) 100% Flax

Figure 3 ((a) to (d)) are used to check the fit of the Fick diffusion model on the experimental data. The fit of the experimental values with the Fick model shows an overlay more or less perfect; the fit is almost perfect for stacking sequences [F C F C]_s and [C C F F]_s, less perfect for 100% flax while the gap is increasing more for the [C F C F]_s. It is then obvious that the water recovery can not only be modelling with a unique Fick law. Contrary to composites reinforced by conventional reinforcements where the water sorption phenomenon is controlled by the matrix, in NRFC sorption is also associated with flax fibres because of their hydrophilic character. Moreover the flax ply location plays a major role in the sorption kinetics by their contribution to increasing the degree of porosity. For hybrid carbon-flax composite, the location of flax plies on the surface of the structure exposes them to more moisture and increases the absorption of water. According to table 1, the diffusion coefficient increases when flax plies are placed near the composite external surface instead of the heart. Therefore the diffusion coefficient is nearly doubled when a flax ply is placed at the surface. On the other side, placing the carbon plies on the surface reduces the diffusion coefficient of water but does not stop the water recovery. Indeed, the water content at saturation is nearly constant whatever the hybrid stacking sequence.

4.2. Buckling test response

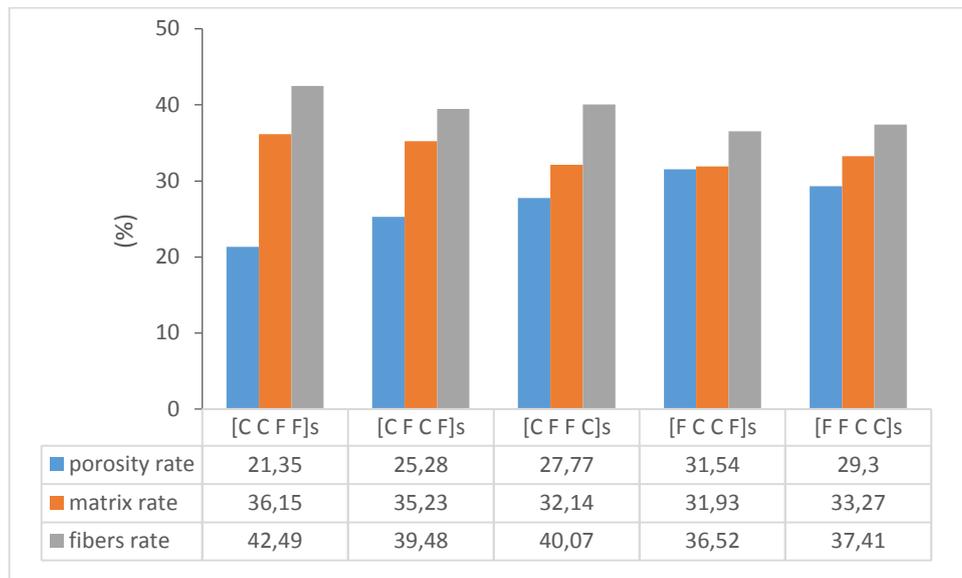


Figure 4. Volume rate of each component (matrix, fibre and porosity) for the different stacking sequences

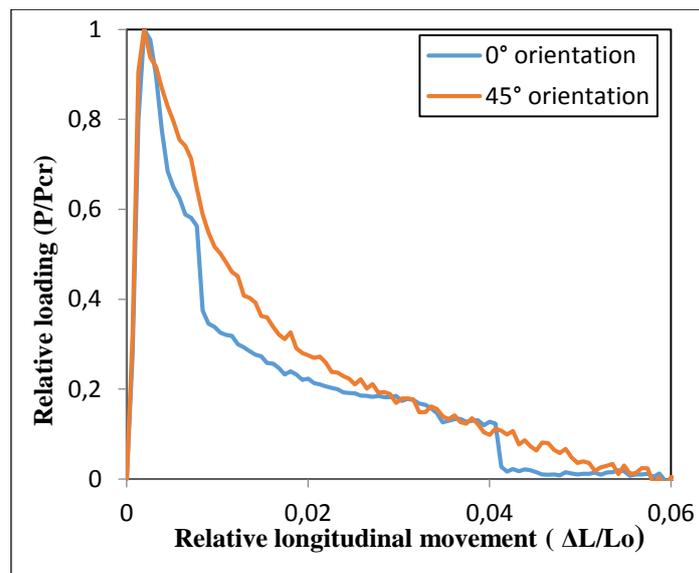


Figure 5. Normalised buckling curve (relative loading $\frac{P}{P_{cr}}$ according to the relative longitudinal movement $\frac{\Delta L}{L}$) for [C F F C]_s specimens cut in the 0 ° and 45° orientations

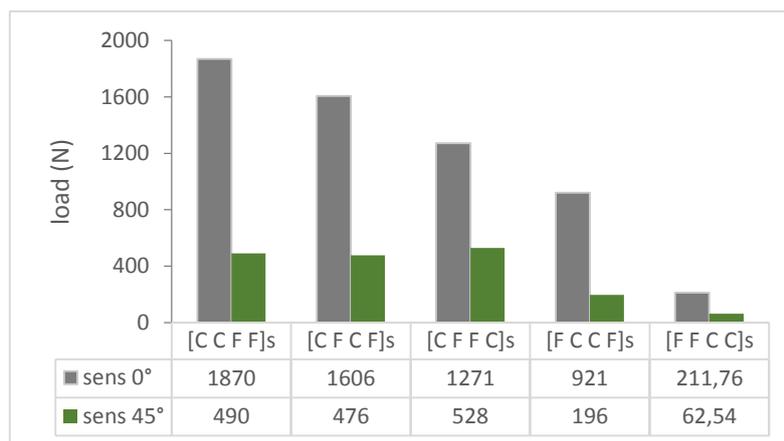


Figure 6. Buckling critical load for the different stacking sequences

Another stacking sequence has been manufactured in this part in order to analyze its response to a buckling test and its damage process. The porosity and matrix rate of the several stacking sequences values very significantly (figure 6), but their total amount is consistent. Indeed, the higher is the porosity rate, the lower is the matrix rate. The more the carbon plies are placed outside the stacking sequence, the lower is the porosity rate. As carbon fibre has a good compatibility with matrix and carbon fibre fabric has a high filling rate, consequently carbon fibre ply placed on surface plate limits the formation of surface porosity.

The shape of the buckling curve is almost the same whatever the specimen stacking sequence (figure 5). After an elastic compressive stage, when the buckling critical load is reached P_{cr} , the sample rigidity collapses suddenly. For the specimen cut in 0° direction, the post-buckling behaviour is characterised by some brutal drops in the curve associated to delamination between composite plies, for the specimen tested in the 45° direction, no delamination is observed.

The buckling critical load depends on the sample stacking sequence (figure 6). The higher value is obtained with the [C C F F]_s, and the buckling critical load decreases when carbon plies are placed in the heart of the sample. Thereby stacking sequences with external carbon plies presents the larger rigidity and consequently the larger critical buckling loads. Moreover as the stacking sequences with external carbon plies have the weakest porosity rate, their Young modulus is higher than material with porosity. By combining these two effects, the composites following the rule of external carbon plies are the more resistant to buckling solicitation.

5. Conclusion

Measurement of porosity rate of the different hybrid stacking sequences, sorption test and buckling test were carried to produce a global result. The measure of porosity rate allows to determine the impact of the position of the flax plies on the porosity rate of the whole structure, we deduce that more carbon plies are near the surface, the less the porosity rate is, which can be explained by the architecture and structure of the flax fabric which is insufficiently filled. Thereafter, a sorption test were conducted to determine suitable sorption mechanism to these plates and adjust it with Fickian modelling and determine water content at saturation and the diffusion coefficient of each stacking sequence. We ended a buckling test to determine the damage process of the various stacking sequences. As a result, we deduce that the buckling critical load depends on the stacking sequence and precisely the position of carbon plies inside the structure as well as the orientation of the fibres compared to the solicitation axis.

References

- [1] C. A. S. Hill and H. P. S. Abdul Khalil. Effect of fibre treatments on mechanical properties of coir or oil palm fibre reinforced polyester composites. *Journal of Applied Polymer Science*, 78:1685-1697, 2015.

- [2] A. Mustapha, Z. Wajdi, S. Hamid, A. Rezak and J.-M. Berthelot. Evaluation of the damping of hybrid carbon–flax reinforced composites. *Composite Structures*, 132:148–154, 2015
- [3] Z. S. Sagheri, I. El Sawi, H. Bougherara and R. Zdero. Biomechanical fatigue analysis of an advanced new carbon fibre/flax/epoxy plate for bone fracture repair using conventional fatigue tests and thermography. *Journal of the Mechanical Behavior of Biomedical Materials*, 35:27–38, 2014
- [4] H. N. Dhakal, Z. Y. Zhang, R. Guthrie, J. MacMullen and N. Bennett. Development of flax/carbon fibre hybrid composites for enhanced properties. *Carbohydrate Polymers*, 96:1-8, 2013.
- [5] B. Madsen and H. Lilholt. Physical and mechanical properties of unidirectional plant fibre composites – an evaluation of the influence of porosity. *Composites Science and Technology*, 63:1265-1272, 2003
- [6] Z.E. Cherif, C .Poilâne, A. Vivet, B. Ben Doudou and J .Chen. About optimal architecture of plant fibre textile composite for mechanical and sorption properties. *Composite Structures*, 140:240-251, 2016.
- [7] Y. Swolfs, L. Gorbatikh and I. Verpoest. Fibre hybridisation in polymer composites: A review. *Composites Part A: Applied Science and Manufacturing*, 67:181-200, 2014.
- [8] M. R. Sanjay, G. R. Arpitha and B. Yogesha. Study on mechanical properties of natural – glass fibre reinforced polymer hybrid composites: a review. *Materials Today: Proceedings*, 2: 2959-2967, 2015.
- [9] M. Assarar, W. Zouari, H. Sabhi, R. Ayad and J.M. Berthelot. Evaluation of the damping of hybrid carbon–flax reinforced composites. *Composite. Structures*, 132:148–154, 2015.
- [10] Z.S. Bagheri, I. El Sawi, H. Bougherara and R. Zdero. Biomechanical fatigue analysis of an advanced new carbon fibre/flax/epoxy plate for bone fracture repair using conventional fatigue tests and thermography. *Journal of the Mechanical Behavior of Biomedical Materials*, 35:27–38, 2014.
- [11] H.N. Dhakal, Z.Y. Zhang, R. Guthrie, J. MacMullen and N. Bennett. Development of flax/carbon fibre hybrid composites for enhanced properties. *Carbohydrate Polymers*. 96:1-8, 2013.
- [12] C.H. Shen, G.S. Springer. Moisture absorption and desorption of composite materials, *Journal of composite materials*, 10:2-20, 1976.