INFLUENCE OF WATER ON THE CYCLIC BEHAVIOUR OF A WOVEN GLASS/PA6,6 COMPOSITE

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Abstract
This study aims to analyze the influence of water on the mechanical behavior of a woven thermoplastic composite. Both tensile static and cyclic behaviors are investigated, for three conditionings: RH0, RH50 and RH100. For a better understanding, three textile layups have been used, namely $[(0/90)_3]$, $[(90/0)_3]$ and $[(\pm45)_3]$. Static tests have highlighted the plasticizing effect of water on the polyamide 6,6 matrix. This result was supported by DSC measurements, since it has been shown that the glass transition temperature decreases with the moisture content. Tension-tension fatigue tests have shown that for highest fatigue lives, the influence of moisture is almost negligible. The final objective of this study is to model the fatigue life of the composite for all studied combinations of moisture content and layup orientation. Thus, an existing fatigue life model, previously developed by Eparaachchi et al. [1], have been improved to fit this requirement. This model has shown a good fitting to experimental data for all moisture content / layup configurations.

1. Introduction

The automotive industry shows increasing interest for continuous glass fibre composites with thermoplastic matrix. The development of this type of material is mainly due to their ease of design, light weight, low cost, recyclability and good mechanical properties [2–4]. Even if thermoplastic polymers provide the advantage to be recyclable, some of them present the major drawback to be sensitive to water [4–7]. It is the case of PA6,6 [8–10], mostly used in automotive structural parts. During their service life, these parts undergo cyclic loading in various moisture conditions. It is thus important to be able to predict the fatigue life of this type of composite for design purpose.

As a starting point, the influence of moisture content on thermal and mechanical properties of the glass fibre reinforced PA6,6 composite (GFRPA66) is studied. Then, the tension-tension fatigue behaviour of the material studied is investigated for the three moisture contents and the three layup orientations chosen. These tests are supplemented with fatigue tests in a climatic chamber, allowing the regulation of both the test temperature and moisture. Finally, a fatigue life model was developed in order to predict the fatigue life of the GFRPA66 in any moisture/layup configuration.

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2. Material and Methods

2.1. Tested Material

The composite material studied is made of three plies of a 2/2 twill woven glass fabric impregnated with polyamide 6,6 resin. The glass fibre fabric has a weight of 600g/m² and a warp to weft ratio of 50/50. The fibre mass fraction (m_f) is equal to 0.63 and the void content is below 1%. The resulting composite plates are characterised by a density of 1.78 g/cm^3. The material is provided as plates of 1.53 mm thick and coupons are cut using water jet cutting technique. It has been checked that this technique has no significant influence on the material moisture content. The specimen edges were polished to remove mechanical damage caused by the cutting. The influence of fabric orientation on mechanical properties is studied using three different stacking sequences. The first one, referred as [$(0/90)_3$], has the warp direction of each ply oriented at 0° from the tensile axis (x axis). The second one, referred as [$(90/0)_3$], has the weft direction of each ply oriented at 0° from the tensile axis (x axis). Finally, the [$(\pm 45)_3$] has the warp direction of each ply oriented alternately at +45° and -45° from the tensile axis. Neat PA6,6, made of the same grade as the resin of the GFRPA6,6 composite, was used for the measurement of the glass transition temperature.

2.2. Conditionings

Three conditionings were studied in order to evaluate the influence of moisture on the fatigue behaviour of the composite: RH0, RH50 and RH100. Conditioning was done by the material supplier by following the standard ISO 1110. Whereas RH0 corresponds to the dry-as-moulded state of the composite material, RH50 and RH100 were conditioned in a climatic chamber until weight stabilization. The neat PA6,6 resin was conditioned at the laboratory : samples were either dried in an oven at 35°C without humidity monitoring or immersed in 20°C water. These two conditions are, respectively, referred to as “dry” state and “wet” state. The reference state is defined as ambient temperature and humidity (respectively, about 20°C and 48 ± 5% RH).

2.3. Experimental procedure

2.3.1 DSC measurements

Polyamide 6,6 glass transition temperature has been determined using a differential scanning calorimeter (DSC) Q20 from *TA Instruments* on pure resin samples. The glass transition temperature is hard to detect, hence modulated DSC (M-DSC) was used [11]. Modulation amplitude was chosen according to the modulation period and the heating rate in order to stay in “heat only” conditions. In these conditions, the modulation does not imply any cooling phenomenon during the test.

2.3.2 Mechanical testing

Quasi-static tensile tests were performed using an INSTRON 4505 electromechanical machine with a cross-head speed of 1 mm/min, strain being measured by a 25 mm gauge length extensometer.
Fatigue tests were performed by using an INSTRON 8501 servo-hydraulic machine. The jaws of test machine clamp 40 mm of each specimen extremity and 80 grit sand papers were used in the jaws to improve clamping. Constant amplitude loads were applied in a sinusoidal waveform at the frequency of 1 Hz in order to limit self-generated heating in the specimen. The stress ratio (R), i.e. ratio between minimum (σ_{min}) and maximum (σ_{max}) stresses, was equal to 0.1 for all tests.

2.3.3 Fatigue Life Model

The fatigue life model used in this study was proposed by Epaarachchi and Clausen [1] and is presented in Eq. 1 [1].

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$$N_f = \left(1 + \left(\frac{\sigma_u}{\sigma_{\max}} - 1\right)\frac{f^{\beta}}{\alpha(1-R)^{\lambda - R|\sin\theta|}}\left(\frac{\sigma_u}{\sigma_{\max}}\right)^{\lambda - 1 - R|\sin\theta|}\right)^{1/\beta} \tag{1}$$

where N_f is the fatigue life, σ_{max} is the maximum fatigue stress, σ_u is the ultimate tensile strength, f is the frequency, R is the stress ratio and θ is the smallest angle between the loading axis and the fibres. The parameter λ is assumed to be equal to 1.6 according to Epaarachchi et al. [1]. Hence, α and β are the only two material parameters that need to be determined using experimental data. Only one S-N curve for a given stress ratio, frequency and lay-up is necessary to determine these parameters.

3. Results

3.1. Glass transition temperature

Glass transition temperatures (T_g) of reference, wet and dry neat PA6,6 samples have been determined using modulated DSC technique. It is thus possible to separate reversing and nonreversing heat flow: the reversing heat flow contains thermodynamic components like glass transition temperature while the nonreversing heat flow contains kinetic events like crystallisation. Reversing heat flow versus temperature curves of reference, wet and dry samples are presented in Fig.1. Results show that T_g decreases with the moisture content.

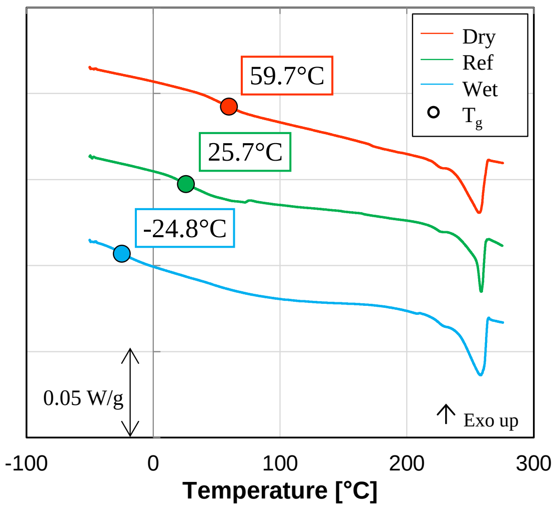

Figure 1. Reversing heat flow from M-DSC thermograms of dry, wet and reference PA-6,6 samples.

3.2. Mechanical properties

Stress-strain curves of both [(0/90)$_3$] and [(±45)$_3$] layup composites are presented in Fig.2. It is worth noting that the [(90/0)$_3$] layup results are not displayed since the stress-strain curves are identical to those of the [(0/90)$_3$] layup. The stress-strain curves for [(0/90)$_3$] composites show a “knee-point” at about 0.4% strain, characteristic of the static behaviour of woven composite [12, 13]. The “knee-point” has been identified as the end of linear part of the stress-strain curve in [(0/90)$_3$] composite.

The stress-strain curves for [(±45)$_3$] is characterised by a short linear part followed by a highly nonlinear part. Mechanical features are summed up in Table 1, between 2 and 6 coupons of each type have been tested. One can notice that the strain at break of [(±45)$_3$] layup is 10 times higher than the strain at break measured for the [(0/90)$_3$] composite. Moreover, results show that mechanical properties are clearly affected by the moisture content.

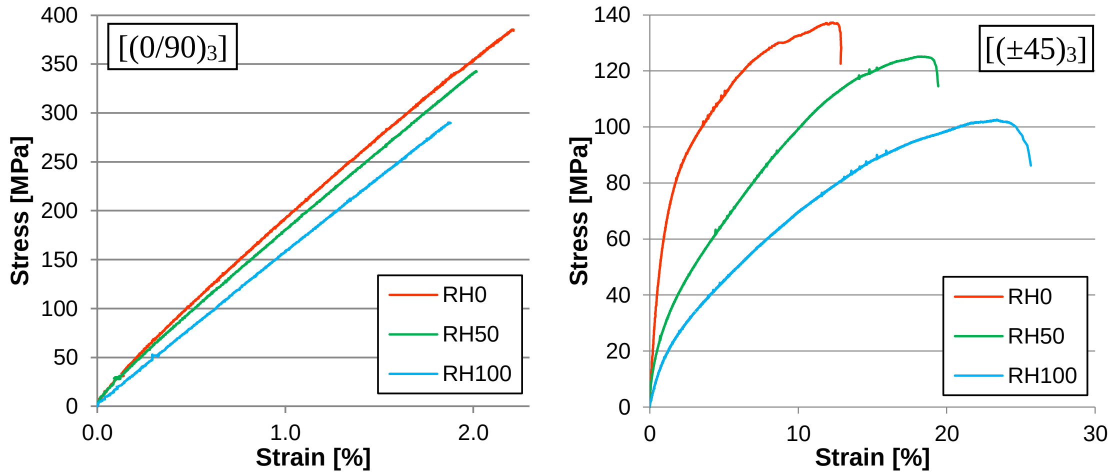

Figure 2. Typical stress-strain curves of [(0/90)$_3$] and [(±45)$_3$] composites at RH0, RH50 and RH100.

Table 1. Tensile properties of [(0/90)$_3$] and [(±45)$_3$] GFRPA66 composites at RH0, RH50 and RH100.

	[(0/90)$_3$] and [(90/0)$_3$]			[(±45)$_3$]		
	RH0	RH50	RH100	RH0	RH50	RH100
σ_u [MPa]	377.7 ± 11.0	343.5 ± 0.7	290 ± 0.5	143 ± 8.1	124.4 ± 1.1	102.3 ± 0.3
ε_u [%]	2.15 ± 0.06	2.05 ± 0.04	1.84 ± 0.10	13.30 ± 0.70	18.45 ± 1.39	24.10 ± 2.30
E [GPa]	19.6 ± 0.4	17.5 ± 0.5	16.2± 0.8	6.8 ± 0.8	2.6 ± 0.1	1.3 ± 0.1

3.3. Fatigue life

3.3.1. Fatigue life at ambient temperature and hygrometry

Fatigue tests were performed on a GFRPA66 at a frequency of 1Hz and a stress ratio (R) of 0.1 until failure of coupons (Fig.3). Samples that did not fail after 10^6 cycles were stopped and black horizontal arrowheads were added in Fig.3. The dispersion of fatigue results was not found significant (less than a decade) in light of fatigue tests performed on three coupons at 100 MPa and 80 MPa for the following conditions: [(±45)$_3$] layup and RH0 (Fig. 3a). The little scattering may be a direct consequence of the quality of the material, and particularly its slight porosity (<1%). Both [(0/90)$_3$] and [(90/0)$_3$] layups show the same fatigue behavior (see Fig. 3(b)), mainly because of the presence of glass fibres aligned with the loading axis.

For the highest stress levels: the higher the moisture content, the shorter the fatigue life. This trend is no longer true when it comes to lower stress levels. From about 10^4 cycles, the three S-N curves tend to get closer until experimental data align along only one S-N curve, regardless the moisture condition. The first hypothesis proposed was that the moisture content would change during fatigue tests of several hours at ambient hygrometry. For instance, samples at RH100 might dry during fatigue tests

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and so, the RH100 S-N curve could get closer to the RH50 S-N curve. Thus, to confirm or infirm this theory, fatigue tests were reproduced in a climatic chamber, with hygrometry regulation.

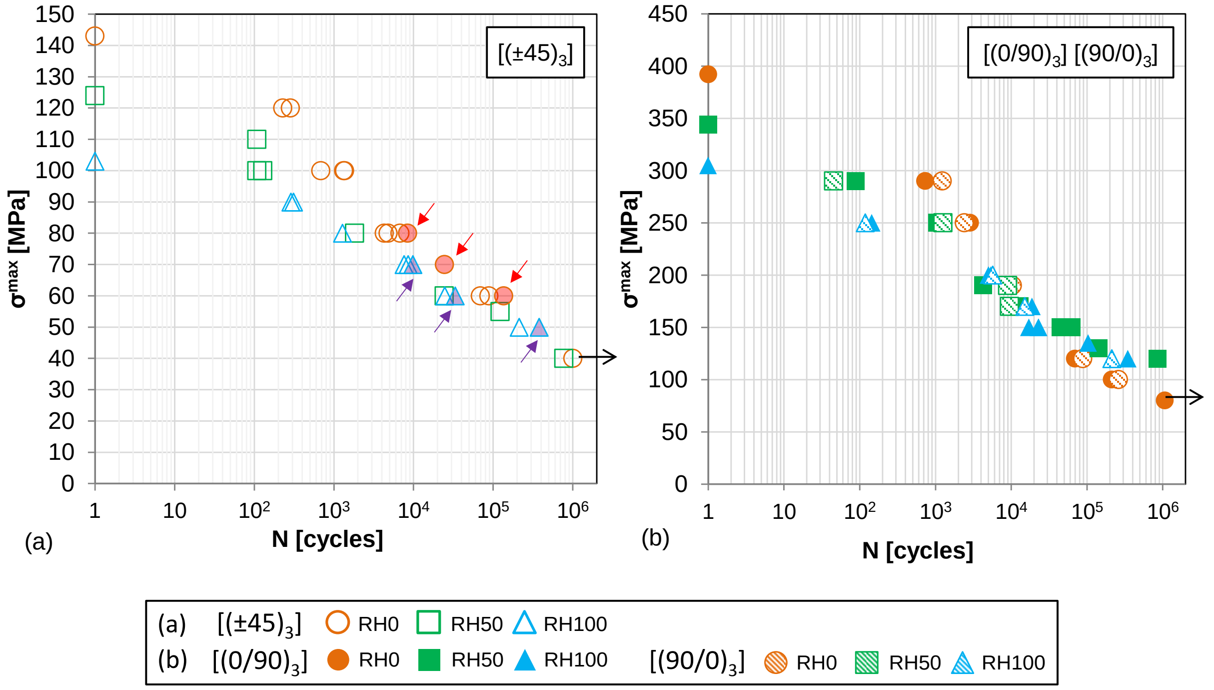

Figure 3. S-N curves of the woven glass fibre / PA6,6 composite for the three moisture conditions (RH0, RH50, RH100) for (a) [(±45)$_3$] layup and (b) [(0/90)$_3$] and [(90/0)$_3$] layups

3.3.2. Fatigue life in climatic chamber

Assuming that RH100 samples tested at room hygrometry tend to dry whereas RH0 samples tend to wet during fatigue tests, fatigue lives presented in Fig. 3 could be respectively maximized and minimized. Therefore, in order to study the influence of the testing moisture conditions, three RH100 samples were tested in a climatic chamber at 20°C and 95% of relative humidity for three different load levels. Similarly, three RH0 samples were tested in a climatic chamber at 20°C and 15% of relative humidity for three different load levels. Results are presented in Fig. 3a with symbols indicated by a colored arrow. Fatigue lives of samples studied in climatic chamber are slightly higher in every case. However, this increase is not significant and is in the dispersion range. Finally, these results indicate that the moisture condition during fatigue test does not really influence the fatigue life. Thus, fatigue lives determined at ambient temperature and hygrometry, presented in Fig. 3, are relevant in the context of this study. To conclude, the fact that the three S-N curves get closer for long fatigue lives is not due to the change in the moisture content in the sample.

4. Discussion

4.1. Plasticizing effect of water

It was shown that the moisture content influences the value of the T_g of the GFRPA66 (Fig. 1). This is a direct consequence of the plasticizing effect of water on polyamide 6,6: water molecule suppresses some hydrogen bondings between amide groups, leading to higher mobility of polyamide chains and thus, to the modification of T_g [14].

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The weakening of the hydrogen bonding in polyamides also leads to the overall reduction of the mechanical properties of the composite (Fig. 2). The $[(\pm45)_3]$ layup having no fibre aligned with the loading axis, the failure mechanisms are thus driven by the PA6,6 matrix itself. The influence of moisture content is then substantial : the Young modulus and stress at break decrease when moisture content increases whereas the strain at break increases. At the opposite, the failure of the $[(0/90)_3]$ layup is mainly driven by the fibre behaviour, insensitive to moisture variation. Thus, from the linear part of the curve, the calculated Young modulus is not clearly affected by the presence of water. However, degradation of interfaces still leads to the reduction of strain and stress at failure.
Results show that the tensile strength of the material is linearly dependent of the relative humidity content in the sample and can be modelled by Eq. 2.

$$\sigma_u^{\theta} = \sigma_u^{\theta,RH50} - a_{\theta} \cdot (RH - 50) \tag{2}$$

where RH is the relative humidity percentage, σ_u^{θ} is the tensile strength for a given layup, referred as θ here. $\sigma_u^{\theta,RH50}$ is the tensile strength for a given layup at RH50 and a_θ is the slope of the linear model. Values of the latter are given in Table 2. This simple linear model allows the determination of the tensile strength of the material, for a given layup, for any relative humidity content.

Table 2. Values of $\sigma_u^{\theta,RH50}$ and a_θ for the two layups.

θ	$\sigma_u^{\theta,RH50}$ [MPa]	a_θ
$[(0/90)_3]$ and $[(90/0)_3]$	343.5	0.88
$[(\pm45)_3]$	124.4	0.41

4.2. Fatigue life modelling

The fatigue life model used in this study has been presented in section 2.3.3. The determination of the two parameters α and β is presented elsewhere [17]. This study suggests an enhancement of this model, by including the relative humidity. By combining Eq. 1 and Eq. 2, it is possible to take into account the evolution of the stress at failure depending on the moisture content (Eq. 3).

$$N_f = \left(1 + \left(\frac{\sigma_u^{\theta,RH50} - a_\theta \cdot (RH-50)}{\sigma_{\max}} - 1 \right) \frac{f^{\beta}}{\alpha(1-R)^{1.6-R|\sin\theta|}} \left(\frac{\sigma_u^{\theta,RH50} - a_\theta \cdot (RH-50)}{\sigma_{\max}} \right)^{0.6-R|\sin\theta|} \right)^{1/\beta} \tag{3}$$

As a result, the only experimental data necessary to apply this model are: the tensile strength of the material for a given θ, at RH50 ($\sigma_u^{\theta,RH50}$), the value of a_θ and one S-N curve for the determination of α and β (the configuration RH50, $[(0/90)_3]$ layup has been chosen here). Then, the fatigue lives can be determined for any moisture percentage, stress level, stress ratio and frequency.
Fatigue lives calculated with the enhanced model proposed (Eq. 3) are given in Fig. 5. The model estimates quite well the fatigue life of the material in all cases. With the exception of $[(0/90)_3]$ and $[(90/0)_3]$ layups at RH0, the model is conservative and captures the fatigue behavior of the GFRPA66 composite.

5. Conclusion

This study discusses the influence of the moisture content on the static and fatigue behavior of a woven glass-fibre-reinforced composite with polyamide 6,6 matrix. Three layups were studied,

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namely [$(0/90)_3$], [$(90/0)_3$] and [$(\pm45)_3$] as well as three moisture conditions, referred as RH0, RH50 and RH100. Fatigue tests were performed on the different possible configurations (layup / moisture condition) both in air environment and climatic chamber. Experimental results were used to develop a semi-empirical model. This model succeeded in providing an enhanced model, taking into account the moisture content.

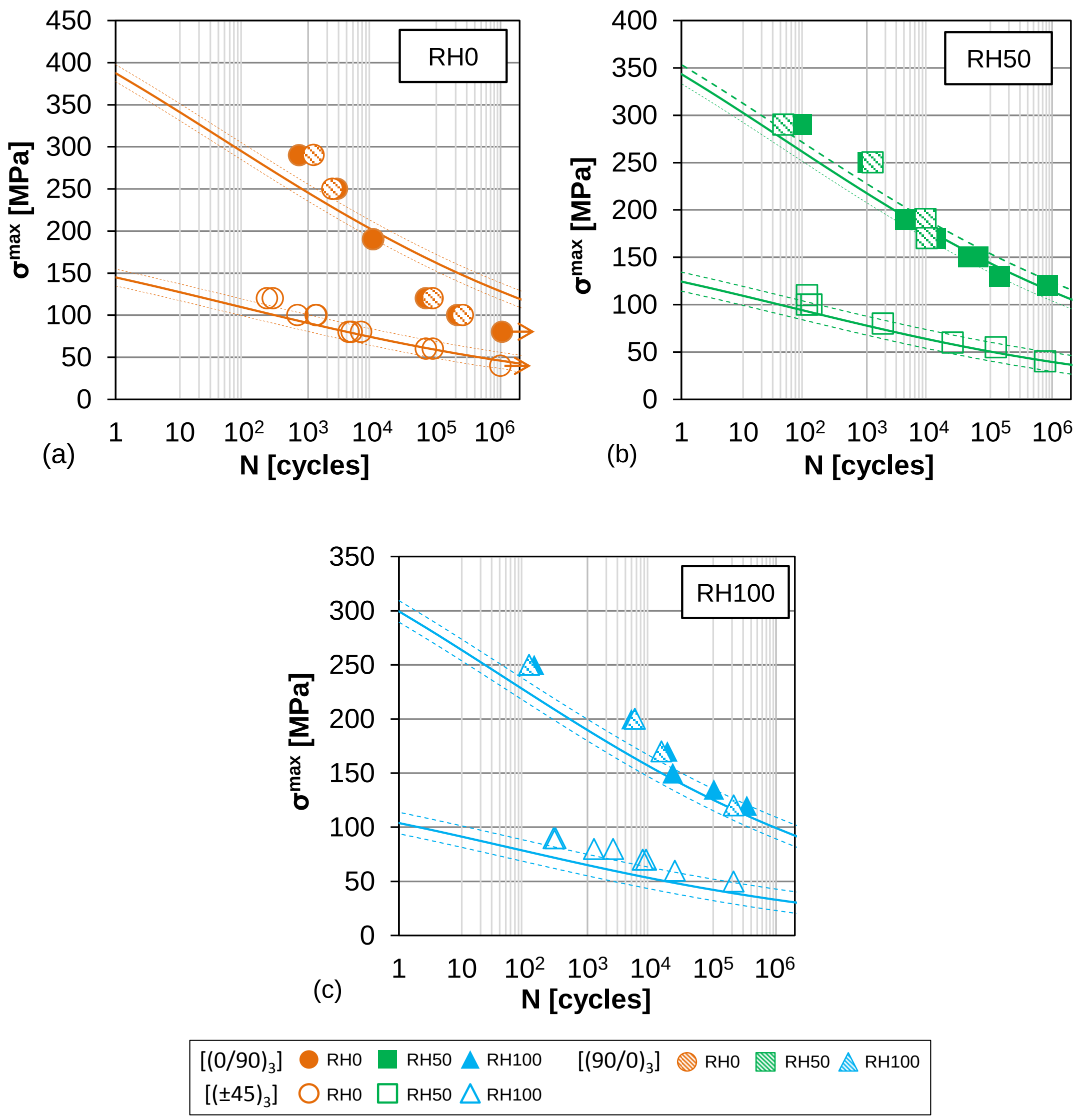

Figure 5. Experimental and calculated fatigue lives of [$(0/90)_3$], [$(90/0)_3$] and [$(\pm45)_3$] layups of woven glass fibre / PA6,6 composite for (a) RH0, (b) RH50 and (c) RH100

References

[1] J. A. Epaarachchi and P. D. Clausen, “An empirical model for fatigue behavior prediction of glass fibre-reinforced plastic composites for various stress ratios and test frequencies,” *Compos. Part A Appl. Sci. Manuf.*, vol. 34, pp. 313–326, 2003.

[2] A. Launay, Y. Marco, M. H. Maitournam, I. Raoult, and F. Szmytka, “Cyclic behavior of short glass fiber reinforced polyamide for fatigue life prediction of automotive components,”

Procedia Eng., vol. 2, no. 1, pp. 901–910, Apr. 2010.

[3] S. Barré and M. L. Benzeggagh, “On the use of acoustic emission to investigate damage mechanisms in glass-fibre-reinforced polypropylene,” *Compos. Sci. Technol.*, vol. 3538, no. 94, pp. 369–376, 1994.

[4] M. F. Arif, F. Meraghni, Y. Chemisky, N. Despringre, and G. Robert, “In situ damage mechanisms investigation of PA66/GF30 composite: Effect of relative humidity,” *Compos. Part B Eng.*, vol. 58, pp. 487–495, Mar. 2014.

[5] M. P. Foulc, a. Bergeret, L. Ferry, P. Ienny, and a. Crespy, “Study of hygrothermal ageing of glass fibre reinforced PET composites,” *Polym. Degrad. Stab.*, vol. 89, no. 3, pp. 461–470, Sep. 2005.

[6] A. Boubakri, K. Elleuch, N. Guermazi, and H. F. Ayedi, “Investigations on hygrothermal aging of thermoplastic polyurethane material,” *Mater. Des.*, vol. 30, no. 10, pp. 3958–3965, Dec. 2009.

[7] L. Gautier, B. Mortaigne, and V. Bellenger, “Interface damage study of hydrothermally aged glass- fibre-reinforced polyester composites,” *Compos. Sci. Technol.*, vol. 59, pp. 2329–2337, 1999.

[8] M. Broudin, V. Le Saux, P. Y. Le Gac, C. Champy, G. Robert, P. Charrier, and Y. Marco, “Moisture sorption in polyamide 6.6: Experimental investigation and comparison to four physical-based models,” *Polym. Test.*, vol. 43, pp. 10–20, 2015.

[9] A. Benaarbia, A. Chrysochoos, and G. Robert, “Influence of relative humidity and loading frequency on the PA6.6 thermomechanical cyclic behavior: Part I. mechanical and thermal aspects,” *Polym. Test.*, vol. 40, pp. 290–298, 2014.

[10] N. Jia and V. A. Kagan, “Mechanical Performance of Polyamides with Influence of Moisture and Temperature – Accurate Evaluation and Better Understanding,” *Plast. Fail. Anal. Prev.*, pp. 95–104, 2001.

[11] A. Malpot, F. Touchard, and S. Bergamo, “Effect of relative humidity on mechanical properties of a woven thermoplastic composite for automotive application,” *Polym. Test.*, vol. 48, pp. 160–168, 2015.

[12] T. Osada, A. Nakai, and H. Hamada, “Initial fracture behavior of satin woven fabric composites,” *Compos. Struct.*, vol. 61, no. 4, pp. 333–339, Sep. 2003.

[13] H. Ryou, K. Chung, and W. Yu, “Constitutive modeling of woven composites considering asymmetric / anisotropic , rate dependent , and nonlinear behavior,” *Compos. Part A Appl. Sci. Manuf.*, vol. 38, pp. 2500–2510, 2007.

[14] S. L. Angioni, M. Meo, and A. Foreman, “A critical review of homogenization methods for 2D woven composites,” *J. Reinf. Plast. Compos.*, vol. 30, no. 22, pp. 1895–1906, 2011.

[15] C. J. Jones, R. F. Dickson, T. Adam, H. Reiter, and B. Harris, “The environmental fatigue behaviour of reinforced plastics,” *Proc. R. Soc. London. Ser. A*, vol. 396, pp. 315–338, 1984.

[16] Y. Hu, A. W. Lang, X. Li, and S. R. Nutt, “Hygrothermal aging effects on fatigue of glass fiber / polydicyclopentadiene composites,” *Polym. Degrad. Stab.*, vol. 110, pp. 464–472, 2014.

[17] A. Malpot, F. Touchard, and S. Bergamo, “Influence of moisture on the fatigue behaviour of a woven thermoplastic composite used for automotive application,” *Mater. Des.*, vol. 98, pp. 12–19, 2016.

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