BEHAVIOUR OF BOLTED COMPOSITE JOINTS IN HYGRO-THERMAL ENVIRONMENTS

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Abstract

Environmental conditions have an important impact on the physical and mechanical properties of polyamide 6 used as a matrix in the studied thermoplastic composite material. The controlled desorption and absorption procedures define the time needed to provide the moisture content for required relative humidity levels. Then the campaign of tensile testing reveals the influence of varying parameters on the in-plane properties of the composite material. They are related to both the testing environment, represented by temperature and moisture content in the material, and fibre orientation in order to access the fibre and resin-dominated properties that vary due to environmental conditions. The glass transition temperature is evaluated for all relative humidity levels under consideration in order to analyse the material state as a result of exposure to environmental conditions. Consequently, the out-of-plane behaviour also changes, which may lead to rapid loss of preload in case of bolted joints of the composite material. To avoid the damage of tows during bolt tightening, the preliminary contact compression tests of the composite circular specimens are performed.

1. Introduction

The use of composite materials is continuously increasing in various domains. Their intensive investigation began in the last century, which led to the numerous applications and the replacement of metallic components. Polyamides are engineering thermoplastic materials, also widely used as resins in composite materials. Depending on the polymerization type, one can distinguish several grades of polyamides, thus the current paper is focused on the polyamide 6 reinforced with continuous glass fibres. However, this material is highly sensitive to environmental conditions [1-6] due to the matrix that possesses amide groups. Hence, the composite material undergoes the plasticization leading to a significant physical degradation when is in service [3], [6-7]. The second type of degradation is chemical, resulting in hydrolysis process. Prolonged immersion in water at high temperatures provokes the chain scission of polymer inducing the irreversible decrease of mechanical properties [8-9]. Nevertheless, this degradation type is not considered in this work.
Polyamide 6 is a semi-crystalline polymer constituting of amorphous and crystalline phases with the degree of crystallization around 30-40% [1]. Only the amorphous phase can absorb the moisture, which has the effect similar to temperature [10]. Humid environment activates the chain mobility of polymer in the amorphous region, resulting in the important change of properties, which has been described in the literature. Le Gac et al. [7], Ishisaka et al. [10] showed the reduction of glass transition temperature from 66°C with no water content to -10°C when saturated in water. It signifies

that the state of the material, either glassy or rubbery, is defined by the moisture content. According to Taktak et al. [3], Silva et al. [6], Le Gac et al. [7], the Young's modulus, the stress at yield and the ultimate stress decrease for PA6 exposed to a humid environment. Thermoplastic composites are also known for their low resistance to creep. Yang et al. [11] showed that the creep deformation of PA66 filled with nanoparticles was higher at 50°C rather than 23°C due to the T_g close to 50°C. Also, the exposure to 80°C lowered the load bearing capability of specimens and increased the extension of PA66. In addition, the creep rate was dependent on testing temperature and stress level. Chevali et al. [12] reported very low creep compliance of PA66 during flexural creep tests, which was slightly increasing from 23°C to 90°C. However, Lyons [13] demonstrated a strong effect of the temperatures 23°C, 100°C and 150°C on creep rate of polyamides, thus the influence of temperature was similar to its influence on the tensile strength. The bending creep behaviour at different stress levels was also studied by Hadid et al. [14]. He presented the values of creep strains of polyamide 66 reinforced with glass fibres for stresses from 69 MPa till 259 MPa, which increase from the lowest to the highest loading.
According to above-mentioned information, the creep behaviour is highly dependent on the testing temperature, therefore can be also influenced by the humid environment. Hence, the idea of the current work is not to study the creep of material induced by compressive, bending or tensile forces, but to investigate the loss of bolt preloading force, inducing the compressive load in out-of-plane direction, in terms of varying parameters of specimens made of the thermoplastic composite.

2. Methods and materials

2.1. Creep

The composite materials can deform while subjected to the stresses below the matrix elasticity yield point. This physical phenomenon is named creep. The deformations after the yield point, caused by the creep phenomena, are irreversible; however, they are not immediate and result from a long-time exposure to mechanical stresses. Environmental parameters, such as temperature and humidity, lead to the sudden drop of mechanical properties and also increase the creep deformation and its rate [11-13]. Therefore, the duration of exposure to the high stress, temperature and humidity induces bigger strains, which, in its turn, emphasizes the viscous nature of a material. Composites, assembled with bolts, sustain a high level of preload in order to avoid opening of interfaces of tightened parts [15] and to insure fatigue endurance of fasteners. Hence, out-of-plane creep deformation, caused by constantly applied compressive forces, may significantly affect the pre-tightening procedure of thermoplastic composite joints, which can lead to non-controlled consequences [16].

2.2. Materials and tools

The studied material is polyamide 6 reinforced with continuous roving glass fibres of a twill weave 2×2, supplied by Lanxess. It was delivered in form of panels of 2 mm, 4 mm and 6 mm thicknesses with 0.5 mm thickness per layer. Its density is 1.8 g.cm^3 and fibre volume content is 47%. The circular samples of diameters 36 mm and 18 mm were cut with a water jet. All the samples have a through-hole of 13 mm for bolts, located in the middle. Hence, there are 6 different geometrical configurations to be tested. The other type of specimens, also cut with a water jet and used for tensile testing, is rectangular of 250 mm×25 mm×2 mm.
Bolts and nuts, made of high-tensile stainless steel with a good resistance to high and low temperatures, are provided by Bumax company. Bolts were instrumented with 2 uniaxial strain gauges diametrically opposed. In addition, the calibration of bolts was done till 60 kN in order to avoid the plasticization. This procedure allows the direct conversion of bolt deformation, caused by tensile load, into force, which is vital for the proper experiments.

2.3. Desorption and humid ageing

The initial moisture content in received laminates was unknown. Therefore, before proceeding with the absorption, all the samples were dried at 90°C in a vacuum that significantly accelerates desorption, compared to ambient temperature, without damaging the material. The evolution of moisture content is evaluated using the gravimetric method [1-3]. It consists in weighing a sample on a regular basis, using a high-precision balance of 0.1 mg. Consequently, comparing the initial mass of a sample at the beginning of desorption and at several time-steps, we obtain the mass change due to loss of moisture (see Eq. 1). For each geometrical configuration, diameter and thickness, 3 samples are used in order to reveal consistent results and avoid probable errors of measurements.

$$C_{glob} = (M_0 - M_t)/M_0. \tag{1}$$

In the equation above M_0 represents the initial mass of a sample, M_t is the mass of a sample at the time-step t and C_{glob} is the moisture content at the time-step t. This method is widely used in the literature for the reason of its simplicity and easy application. Nevertheless, it has one major disadvantage, which consists in taking samples out of the environmental chamber for weighing. However, the time during which they are out is barely 5 minutes, thus the mass alteration can be considered as negligible. Once the loss of moisture content reached the threshold and remains stable, we assume that the material is in “dry-as-moulded” condition where Relative Humidity (RH) level is about 0%. In other words, samples do not contain any moisture, or its amount is very little and unimportant. This is the first condition we are interested in. Also, it allowed us to estimate real time to bring the material to minimal moisture content.
The second and third conditions that we chose to analyse are 2 moisture contents corresponding to the humid ageing in RH 50% and RH 85%. For this purpose, samples are exposed to the environment, controlled in humidity and temperature, during some time. The temperature in the environmental chamber is kept constant and remains equivalent to desorption process, so 90°C, in order to preserve consistency of all the parameters. Hence, we are capable to evaluate the time required to bring the material to needed moisture contents corresponding to 50% and 85% of RH by using the same gravimetric method (Eq. 1). It is important to mention that no chemical damage occurs during the process [9].

2.4. Evaluation of glass transition temperature T_g

The temperature at which the transition occurs from glass to the rubbery state for amorphous part of a thermoplastic like polyamide 6, is 60°C according to the material supplier, and also proved by literature review [7], [10]. It is relevant to mention that it is valid only for material in RH0 condition. Hence, the glass transition temperature remains of great importance and is evaluated using the Q800 Dynamical Mechanical Analyser. Three samples per each RH level are loaded with the 3-point bending method in the range of temperatures from -50°C till 150°C. The sample geometry is 60 mm×10 mm×2 mm.

2.5. In-plane tensile testing

An experimental campaign of tensile testing was carried out in order to study the whole range of mechanical properties as a function of environmental conditions. The latter is introduced in terms of temperature and level of RH. As already stated, humid environment causes a significant decrease, impacting, in its turn, mechanical properties [3], [6-7]. The importance of these experiments consists in obtaining data of fibre-dominated, resin-dominated composite material and the neat matrix, exposed to negative and positive temperatures during testing. Samples, used for testing are [45] angle-ply woven laminates, and have a rectangular shape of 250 mm×25 mm×2 mm and those of [0] and [90]-oriented fibres have dumbbell structure with the smallest width of 25 mm, length 250 mm and thickness 2 mm. All of the samples, 5 per condition, were previously dried and aged in a climatic chamber either only under vacuum for no moisture content or also at RH50/RH85 to obtain the moisture content corresponding to these humidity levels and then tested on Instron 5584 with the loading capacity of 100 kN. Thus, received experimental results with the relation to T_g will allow better comprehension of other properties, like out-of-plane behaviour, resistance to creep and loss of preload, which is crucial in the case of bolted assemblies.

2.6. Out-of-plane contact damage

The material aged at RH 85% undergoes the smallest mechanical forces (see Fig. 6) as compared to RH 0% and 50% due to its viscous nature; consequently, this case is the most critical for the other mechanical charges. Therefore, when compressing 2 parts, a composite circular sample and a metallic part of smaller diameter, contact damage can occur. Visually, it is possible to notice it with the thickness change within the area of the contact of the metallic part as a result of localized plastic deformation after the application of high pressure. Resin out-of-plane compression is neglected in this work, whereas the onset of fibre damage is crucial to define the maximal applicable compressive force. Consequently, since fibres maintain applied force, it is proposed to load samples till the appearance of cracks in tows, hence this approach is adopted in the work. The composite specimens with the diameter of 36 mm and a metallic washer of 18 mm were compressed on the Instron 5584.

3. Results and discussions

3.1. Desorption and humid ageing

Prior to absorption test, the desorption process was necessary to control the initial mass of the samples. The applied gravimetric method is simple, however, it requires some time [1-3]. In order to ensure the repeatability and consistency of results, 3 specimens of 2 diameters were used for each thickness. During preliminary study of desorption on rectangular specimen, it was necessary to choose the temperature to accelerate the drying process without affecting the mechanical properties. For this purpose, the specimens were dried at 50°C, 70°C and 90°C in a vacuum till the constant value of moisture content, C_{glob} (Fig. 1). Conducted in-plane tensile tests in shear mode showed no important influence of the drying temperature, thus allowing us selecting 90°C for the desorption and absorption of moisture and saving around 3 weeks as compared to 50°C.

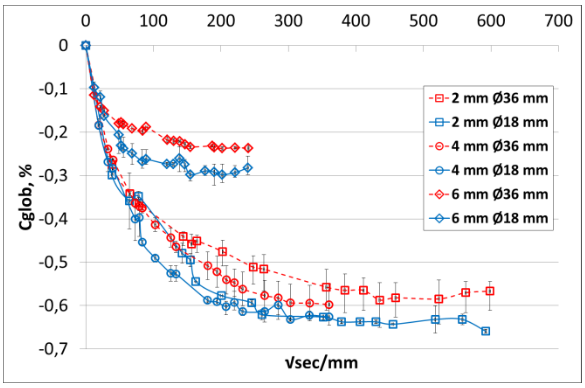


Figure 1. Loss of moisture as a function of the square root of time divided by the specimen thickness.

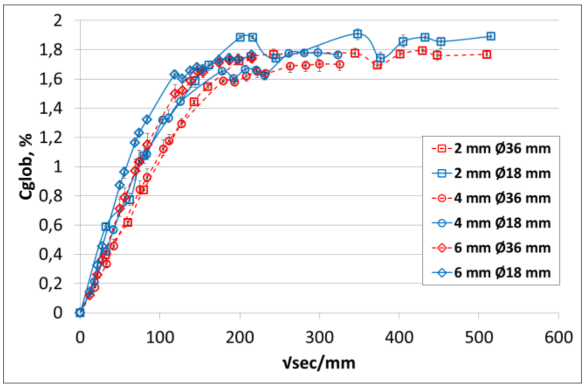


Figure 2. Moisture content as a function of the square root of time divided by the specimen thickness.

In Fig. 1 one can notice the constant level of C_{glob}, which means the dry-as-moulded condition of specimens, or RH0. Hence, theoretically material does not contain water clusters which could leave the amorphous phase without the chemical degradation. The samples of 2 mm and 4 mm have coherent desorption kinetics, whereas for the samples of 6 mm it seems to be different. It can be explained by the initial state of composite laminates. Those of 2 mm and 4 mm were manufactured

many months ago and then stocked in uncontrolled environmental conditions. It led to the very similar moisture content before desorption. The laminate of 6 mm was manufactured 2.5 months before the beginning of the desorption test, which signifies that it was not in the same initial state.
The results of humid ageing of circular composite specimens are introduced in Fig. 2 as a function of ageing time divided by the specimen thickness. Specimens were placed into a climatic chamber at constant RH85 and 90°C. Therefore, all the specimens have similar final moisture content.
When the specimens are large and long enough, the penetration of molecules through the sides can be neglected. For circular samples of 2 mm and rectangular samples of 250 mm×25 mm×2 mm the absorption time remains the same, supposing that in case of samples with the diameter of 18 mm and 36 mm the penetration of water molecules occurs also through the thickness, neglecting the sides. It explains the longer duration of absorption of thicker specimens (Fig. 3), for instance, specimens of 6 mm require the minimal duration of absorption of 17 days, whereas those of 2 mm need only 3 days. Consequently, the moisture absorption rate is higher for thinner composite, and the constant level of C_{glob} is reached faster. This was also reported by Obeid [2].

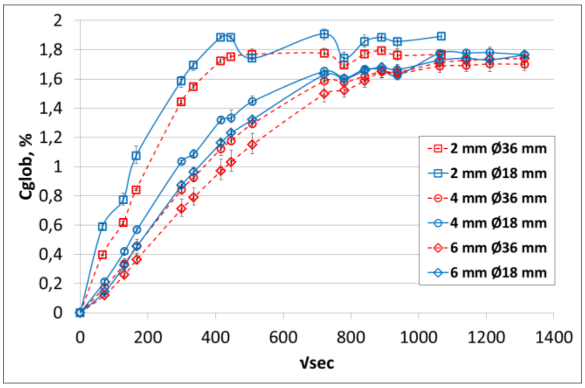


Figure 3. Moisture content as a function of the square root of time.

As stated by Le Gac et al. [7] and Ishisaka et al. [10], the glass transition temperature varies with the moisture content in polyamide 6. Accounting for 3 levels of RH that we want to test, knowing the T_g of the composite is crucial for the analysis of further mechanical response. The importance consists in the varying state of the material, like rubbery when the temperature is above the T_g and glassy when below, which absolutely changes the physical and mechanical characteristics. In order to evaluate the T_g of dry-as-moulded and aged specimens, the dynamic mechanical analysis was applied, where the rectangular-shaped samples were loaded in the bending mode. Obtained results are coherent to the literature, for instance, 57.2°C for RH0 state (see Table 1). In case of the moisture content corresponding to RH50 and RH85, the T_g is 23°C and -1°C respectively.

Table 1. Glass transition temperature of PA6 GF aged at different RH levels.

	HR0	*HR50*	*HR85*	Type of analysis		Specimen
Glass transition temperature	57,2°C	23°C	-1°C	DMA	Bending mode	60 mm×10 mm ×2 mm

3.2. In-plane tensile testing

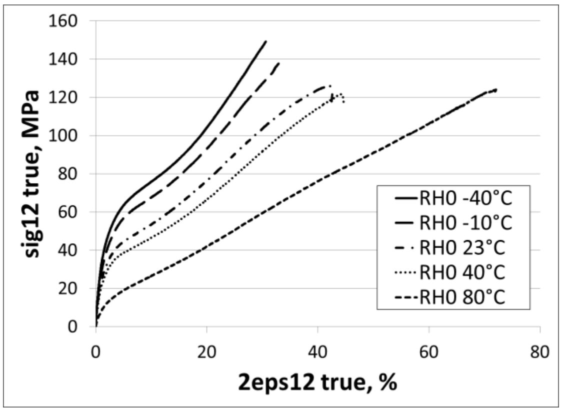
This experimental campaign was an important step towards the evaluation of a wide range of parameters and mechanical properties. Tested at 3 RH levels and 5 temperatures, the behaviour of the composite material indeed depends on the environmental conditions (see Fig. 4 – 6). Mechanical properties are calculated using the data in "true" values, not engineering, due to large strains (Eq. 2 – 4) **[**17]:

$$2\varepsilon_{12\,true} = ln\big(1 + \varepsilon_{xx\,eng}\big) - ln\big(1 + \varepsilon_{yy\,eng}\big) \tag{2}$$

$$\sigma_{xx\,true} = F/S_0 \times (1 + \varepsilon_{xx})^{2\nu^*} \tag{3}$$

$$\nu^* = -\,\dot{\varepsilon}^*_{yy}/\dot{\varepsilon}^*_{xx}. \tag{4}$$

Some obtained results are represented in figures 4 – 6 in terms of true stress-strain curves. The fibre orientation of the specimens is [45] in order to find out the in-plane shear properties that are resin-dominated. High sensitivity to temperature and hydrophilic properties of the amorphous part of polyamide 6 cause the variations of the glass transition temperature, which affects mechanical and physical properties of the resin, consequently the composite [6-7]. Absorbed moisture and temperature play similar roles changing the state from rigid to viscous (Fig. 4 – 6). One can notice that with the increase of both, temperature and moisture content, the ultimate shear stresses and stresses at yield point decrease, as well as the shear modulus, which was also shown by Le Gac et al. [7]. It can be explained by the chain movement when the moisture content is presented and/or the testing temperature is above the T_g. For instance, the glassy state of specimens is well-presented for the tests at the negative temperatures -40°C and -10°C. However, the chain movement leads to higher plastic deformation with the increase of moisture content and/or testing temperature. Also, several tests were carried out for [0] and [90]-oriented specimens. The testing temperature did not show a big impact on the results due to fibre-domination. Yet, the slight increase of the Young's modulus was noticed.



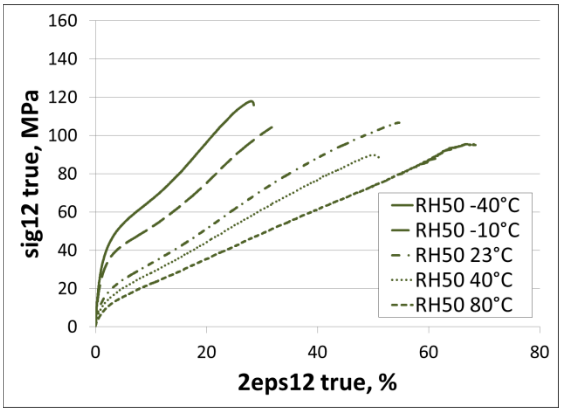


Figure 4. Stress-strain curves of PA6 GF at RH0. **Figure 5.** Stress-strain curves of PA6 GF at RH50.

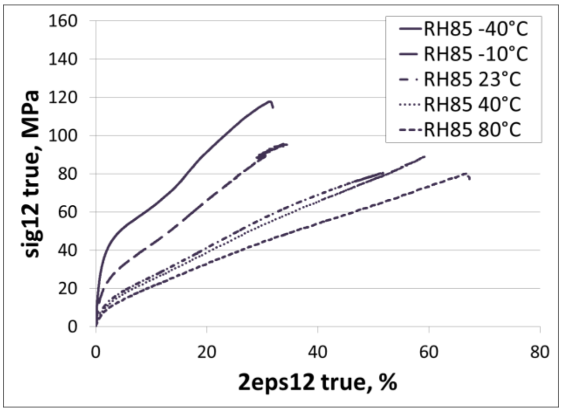


Figure 6. Stress-strain curves of PA6 GF at RH85.

3.3. Out-of-plane contact damage

The out-of-plane damage of material is called contact here due to compression of the metallic and composite materials. It can occur during the preload of bolted joint when the tightening force exceeds the limit of compressive stress of the composite. Proposed by CETIM, the criteria of maximum compressive force consists in non-damaging of fibres. As seen previously, the mechanical behaviour of the composite material is greatly influenced when exposed to RH85. Thus, the samples were previously dried and aged in the humid environment, then tested at the ambient temperature. The first experiment was till the rupture (Fig. 7), thus the splitting path is easily noticeable. The rest of specimens were loaded between 50 and 35 kN with the step of 5 kN and stopped to detect the appearance of cracks. In Fig. 8 one can see the rupture of tows because the applied force is close to the rupture. For the rest, the loading level did not allow propagating crack in tows. However, the compression of the matrix remains important for 45 kN, decreasing towards 35 kN (Fig. 9 – 11). The last value can be defined as acceptable in terms of non-damaging.

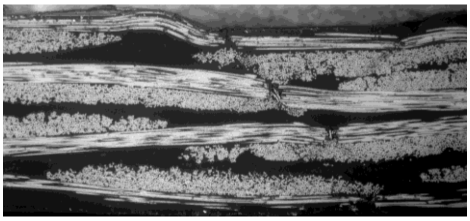


Figure 7. Contact compression till the rupture (≈ 51 kN).

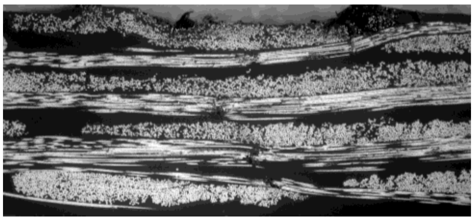


Figure 8. Contact compression till 50 kN.

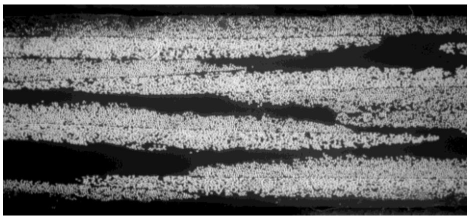


Figure 9. Contact compression till 45 kN

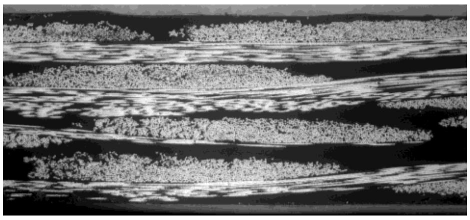


Figure 10. Contact compression till 40 kN.

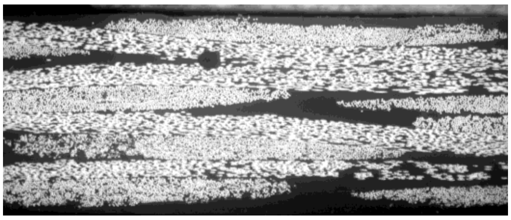


Figure 11. Contact compression till 35 kN.

4. Conclusions

Present paper deals with the analysis of the mechanical and physical properties of polyamide 6 reinforced with glass fibres as a function of environmental conditions that are represented in terms of testing temperature and humid ageing. To obtain the moisture content corresponding to provided RH, desorption procedure was carried out at 90°C in vacuum. Afterwards, the specimens were exposed to the humid environment of RH50 and RH85 at 90°C till the stabilization of moisture content. This process was repeated for the circular specimens.

The change of amorphous state of the matrix occurs as a result of absorbed humidity and testing temperature. This leads to the variation of mechanical properties. The shear modulus, the stress at yield point and ultimate stress reduce with the increase of RH and temperature, whereas the strain grows, showing the viscous nature of the matrix of the composite. Conducted out-of-plane compression tests of the material aged at RH85 allowed selecting the preloading force to avoid contact damaging of the composite in the bolted joint during the experiments. However, some additional experimental results regarding the loss of preload are to be presented during the poster session.

References

[1] D. P. N. Vlasveld, J. Groenewold, H. E. N. Bersee, and S. J. Picken, 'Moisture absorption in polyamide-6 silicate nanocomposites and its influence on the mechanical properties', *Polymer (Guildf).*, vol. 46, no. 26, pp. 12567–12576, 2005.

[2] H. Obeid, 'Durabilité de composites à matrice thermoplastique sous chargement hygro-mécanique: étude multi-physique et multi-échell des relations microstructure -propriétés-états mécaniques', Université de Nantes, 2016.

[3] R. Taktak, N. Guermazi, J. Derbeli, and N. Haddar, 'Effect of hygrothermal aging on the mechanical properties and ductile fracture of polyamide 6: Experimental and numerical approaches', *Eng. Fract. Mech.*, vol. 148, pp. 122–133, 2015.

[4] M. Arhant, P. Y. Le Gac, M. Le Gall, C. Burtin, C. Briançon, and P. Davies, 'Modelling the non Fickian water absorption in polyamide 6', *Polym. Degrad. Stab.*, vol. 133, pp. 404–412, 2016.

[5] A. Krzyzak, J. Gaska, and B. Duleba, 'Water absorption of thermoplastic matrix composites with polyamide 6', *Sci. Journals Marit. Univ. Szczecin*, vol. 33, no. 105, pp. 62–68, 2013.

[6] L. Silva, S. Tognana, and W. Salgueiro, 'Study of the water absorption and its influence on the Young's modulus in a commercial polyamide', *Polym. Test.*, vol. 32, no. 1, pp. 158–164, 2013.

[7] P. Y. Le Gac, M. Arhant, M. Le Gall, and P. Davies, 'Yield stress changes induced by water in polyamide 6: Characterization and modeling', *Polym. Degrad. Stab.*, vol. 137, pp. 272–280, 2017.

[8] C. El-Mazry, O. Correc, and X. Colin, 'A new kinetic model for predicting polyamide 6-6 hydrolysis and its mechanical embrittlement', *Polym. Degrad. Stab.*, vol. 97, no. 6, pp. 1049–1059, 2012.

[9] R. Bernstein and K. T. Gillen, 'Nylon 6.6 accelerating aging studies: II. Long-term thermal-oxidative and hydrolysis results', *Polym. Degrad. Stab.*, vol. 95, no. 9, pp. 1471–1479, 2010.

[10] A. Ishisaka and M. Kawagoe, 'Examination of the time-water content superposition on the dynamic viscoelasticity of moistened polyamide 6 and epoxy', *J. Appl. Polym. Sci.*, vol. 93, no. 2, pp. 560–567, 2004.

[11] J.-L. Yang, Z. Zhang, A. Schlarb et al., 'On the characterization of tensile creep resistance of polyamide 66 nanocomposites. Part I. Experimental results and general discussions', *Polymer*, vol. 47, no. 8, pp. 2791-2801, 2006.

[12] V. S. Chevali, D. R. Dean, G. M. Janowski, 'Flexural creep behavior of discontinuous thermoplastic composites : Non-linear viscoelastic modeling and time-temperature-stress superposition', *Comp. Part A: Appl. Sci. and Manuf.*, vol. 40, no 6-7, pp. 870-877, 2009.

[13] J. S. Lyons, 'Time and temperature effects on the mechanical properties of glass-filled amide-based thermoplastics', *Polym. Test.*, vol. 17, pp. 237-245, 1998.

[14] M. Hadid, S. Rechak, A. Tati, 'Long-term bending creep behavior prediction of injection molded composite using stress-strain correspondence principle', *J. Mat. Sci. and Eng.*, vol. 385, no. 1-2, pp. 54-58, 2004.

[15] NF E25-030-2, 'Fixations - Assemblages vissés à filetage métrique ISO - Partie 2: règles de conception pour les assemblages précontraints - Démarche complète', 2015.

[16] M. Ampleman, 'Performance en fluage des assemblages antiglissement avec des surfaces métallisées dans les ponts en acier', Université Laval, 2016.

[17] Jean Lemaitre, Jean-Louis Chaboche, Ahmed Benallal, Rodrigue Desmorat . Mécanique des matériaux solides - 3ème édition, 2009.