

Durability of mechanical performance of prestressed bolted composite joints in a hygro - thermo - mechanical environment

Ivanna Pivdiablyk

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ECOLE DOCTORALE N° 602 Sciences pour l'Ingénieur Spécialité : Mécanique des solides, des matériaux, des structures et des surfaces

Par

Ivanna PIVDIABLYK

Durability of mechanical performance of prestressed bolted composite joints in a hygro-thermo-mechanical environment

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NOTATIONS

Abbreviations

PA6 GF	Glass-fibre reinforced polyamide 6	
PPS CF	Carbon-fibre reinforced polyphenylene sulphide	
PA6	Neat matrix Polyamide 6	
RH	Relative humidity, %	
DAM	Dry-as-moulded condition or dry material state	
RVE	Representative volume element	
DIC	Digital image correlation	
CTE	Coefficient of thermal expansion	

Reference systems

(1,2,3)	Local frame attached to an elementary ply
(x , y , z)	Global frame attached to a testing machine
$\overrightarrow{b_x}$,	X-axis in global frame (load direction)
$\overrightarrow{b_y}$	Y-axis in global frame (perpendicular to load direction)
$\overrightarrow{b_1}$	X-axis in local frame (warp direction of composite)
$\overrightarrow{b_2}$	Y-axis in local frame (weft direction of composite)

Indices

11	Fibre direction
22	Direction transverse to fibres
33	Through-thickness direction transverse to fibres (out-of-plane)
12	Shear direction in plane 1,2
13	Shear direction in plane 1,3
23	Shear direction in plane 2,3
xx	Longitudinal (load) direction
уу	Transversal (perpendicular to load) direction
ZZ	Through-thickness direction
xy	Shear direction in plane x,y
У	Limit of elastic domain
eng	Mechanical property in "engineering" values
true	Mechanical property in "true" values
max	Maximum value
start	Value at the beginning of experiment
end	Value at the end of experiment
t	Thermal properties
alu	Values of aluminium specimen

M_0	Initial mass of a specimen, g	
M_t	Current mass of the specimen, g	
С	Current water vapour content, g	
C_{max}	Maximum water vapour capacity of the air, g	
C_{glob}	Moisture content, %	
m _{CM}	Mass of composite material, g	
$ ho_m$	Density of matrix, g/cm ³	
$ ho_{CM}$	Density of composite material, g/cm ³	
V_m	Volume of matrix, cm ³	
V _{CM}	Volume of composite material, cm ³	
$\vartheta_{m \ volume}$	Matrix content by volume, %	
ϑ_{mmass}	Matrix content by mass, %	
m_m	Mass of matrix, g	

Symbols related to humid ageing

Symbols related to thermal and hygroscopic expansion

σ_{ij}	Stress tensor
C_{ijkh}	Stiffness tensor
ε_{hk}	Strain tensor
ε^{H}_{hk}	Hygroscopic strain tensor
α_{ii}	Tensor of thermal expansions
β_{ii}^{H}	Tensor of hygroscopic expansions
ΔT_t	Temperature variation
α	Coefficient of thermal expansion, °C-1
T_0	Initial temperature, °C
$k_{T_1 \rightarrow T_2}$	Thermal impact on the grids of the strain gauges between two temperatures T_1 and T_2
ε_{G}^{t}	Thermal strain of gauge grid

Symbols related to mechanical characterisation

F	Tensile force, N
<i>S</i> ₀	Cross-sectional area of a specimen, mm ²
L_0	Initial length of zone of interest of a specimen, mm
W_0	Initial width of zone of interest of a specimen, mm
L	Current length of zone of interest of a specimen, mm
W	Current width of zone of interest of a specimen, mm
T_g	Glass transition temperature, °C
T	Testing temperature, °C
ΔT	Temperature difference between the T_g and the T , °C
Е	Strain
σ	Stress
γ	Shear strain
Ε	Elastic modulus
G	Shear modulus
ν	Poisson's ration
$\dot{arepsilon}^{*}$	Strain rate
ν^*	Ratio of strain rates

heta	Orientation of reinforcement
R^2	Coefficient of determination
y _i and f _i	Data set of measured and estimated values
A and B	Polymer-dependent experimental factors for Kambour slope

Symbols related to homogenisation

$\overline{\sigma}$	Local strain tensor
$\overline{\overline{3}}$	Local stress tensor
$\overline{\overline{\Sigma}}$	Macroscopic stress tensor
Ē	Macroscopic strain tensor
\mathbb{C}	Stiffness tensor
S	Compliance tensor
\mathbb{C}^*	Homogenised stiffness tensor
\mathbb{S}^*	Homogenised compliance tensor
V	Volume of RVE
∂V	Boundary of volume
\overline{u}	Displacement of periodic cell
\bar{x}	Material point
$\bar{\nu}$	Vector of periodic fluctuations

 \bar{n} Normal to the boundary

Symbols related to bolted joints

•	
E_b	Elastic modulus of a bolt, MPa
E_{zzi}	Out-of-plane elastic modulus of a clamped part i in global reference system, MPa
E ₃₃	Out-of-plane elastic modulus of a clamped part in local reference system, MPa
F	Axial force, kN
F_b	Resultant force in a bolt, kN
F _{ext}	External loading, kN
F_0^b	Pretension force in a bolt, kN
λ	Filtering coefficient
β	Load introduction factor
δ_b	Compliance of a bolt, mm/N
δ_{h}^{i}	Compliance of a section i of a bolt, mm/N
$\tilde{\delta_p}$	Compliance of a clamped part, mm/N
δ_{total}	Total compliance of bolted joint, mm/N
φ	Angle of deformation cone
Р	Thread pitch, mm
d	Bolt diameter, mm
d_w	Washer diameter, mm
D_p	External diameter of a clamped part, mm
D_h	Diameter of a clearance hole, mm
d_3	Diameter of engaged internal threads, mm
d_{red}	Diameter of reduced shank, mm
A_n	Nominal bolt cross-section, mm
A_3	Cross-section, associated with d_3 , mm
A_{h}^{i}	Cross-section of a section i of a bolt, mm ²

Α	Cross-section of a clamped part, mm ²				
A_{eq}	Equivalent cross-section of clamped parts, mm ²				
l_b^i	Length of a section <i>i</i> of a bolt, mm				
l _{b head}	Length of not-threaded shank under the bolt head, mm				
l_{red}	Length of reduced shank, mm				
$l_{b \ thread}$	Length of non-threaded shank before threading, mm				
l _{thr shank}	Length of threaded shank except engaged, mm				
L_{pi}	Length of a clamped part <i>i</i> , mm				
L_p	Total length of clamped parts				
L_b	Bolt elongation, µm				
ΔL_b	Increase of bolt elongation at the end of test in comparison to the beginning, %				
\mathcal{E}_G	Bolt deformation from strain gauges, µm/mm				
$\Delta \varepsilon$	Increase of bolt strain at the end in comparison to the beginning, %				
F_{Gi}	Maximum level of applied tension, kN				
μDef	Bolt deformation, µm/mm				
a, b	Parameters of linear equation				
V_r	Volume of interest				

INTRODUCTION

Extensive employment of composite materials, primarily related to the demanding and challenging regulations in automobile and aeronautical industries, leads to the development of modern processing techniques, more accurate computation and modelling approaches, new joining methods. Some joints, for instance, mechanical, are preferred to others despite a large variety of assemblies, mainly developed for composite materials. Thus, the prestressed bolted connections represent an important segment. The conception and computation of bolted joints are regulated by a European standard VDI 2230 with extensions NF E 25030-1 and NF E 25030-2, presenting a complete and a thorough methodology. Nowadays, this document is a reference for the design of preloaded bolted connections. To assist in the conception of metal joints, CETIM has developed and commercialised the software CETIM-Cobra, found on the mentioned standard. The software provides a conception of a bolted joint with preload, based on the properties of elementary constituents of the joint. At present, the software is successively adopted by enterprises in numerous areas of expertise.

Resulting from an intensive application of the mechanical joints partially or entirely integrating composite materials, the design norms possess a significant disadvantage for the reason that they are theoretically developed for metallic materials. Furthermore, the only thermal impact is involved in the computations by employing the coefficient of thermal expansion. Nevertheless, for some composite materials that are integrated into the bolted joints, the thermoplastic matrix and, thus, the composite itself can be sensitive to environmental conditions that include both temperature and humidity. It is, therefore, essential to consider these constraints in order to adapt and improve the software for connections incorporating composite materials in part or entirely. This defines the objective of the research work, namely, to investigate the durability of composite bolted joints, subjected to preload, under different working environmental conditions.

The mechanical behaviour of composite joints has been already studied in a previous PhD thesis, accomplished by R. Hamonou in collaboration of CETIM and GeM (Research Institute in Civil and Mechanical Engineering) [1]. The approach, proposed in this work to determine the compliance of clamped parts, is based on finite element simulations associated with an energy criterion. The results were successfully compared to the standard VDI 2230/ NF E 25030-1/NF E 25030-2. A new analytical model has been developed for a "filtering" coefficient that equally includes a factor of implementation of operational force in terms of clamped part compliance. In the present work, the proposed numerical approach is extended in order to take into consideration the impact of humidity and temperature for the computations of compliance. This method requires the creation of an extensive experimental database for the reason that the work mentioned above has been performed under ambient working environment (assumed to be at a temperature +23°C and a Relative Humidity level of 50%, commonly noted as RH 50%). Besides, the part compliance of thermoplastic composite materials affects the "filtering" coefficient, the prestress level and may vary under the influence of temperature and humidity. Furthermore, the mechanical properties of clamped parts are involved in the characterisation of stresses within the fixation zone during cyclic bolt loading that affects the joint resistance to fatigue. The latter is related to the conservation of preload over some time; however, it is not analysed in the present work. Therefore, an accurate estimation of bolted joint behaviour under different environmental conditions requires a database of mechanical properties of clamped parts.

Prior to the analysis of bolted joints, a thorough characterisation of the given materials – glass-fibre reinforced polyamide 6, carbon-fibre reinforced polyphenylene sulphide and neat polyamide 6 – is necessary, hence, the experimental campaign is proposed at different temperatures and humidity levels. Thereafter,

numerical simulations, based on the finite element method, are implemented in Cast3M at a scale of tow and a Representative Volume Element of the composites. This modelling is intended to enrich the previously developed characterisation protocol and to identify the out-of-plane properties of the composite PA6 GF by the inverse analysis, supplemented by the experimental in-plane data of the neat matrix PA6. This work facilitates the computation of the clamped part compliance as a function of environmental conditions that may serve for a better conception of bolted joints. The last stage consists in the characterisation of preload relaxation over time for different geometries of clamped parts and environmental conditions.

The report is presented in the following order:

The literature review of the studied materials and joints is introduced in Chapter I. It is divided into two sections: the environmental impact on the composite materials and the characterisation of bolted joints. In the first section, the importance of relative humidity, temperature and their global influence on the behaviour is demonstrated. The introduction to hygro-thermal impact on chemical, physical and mechanical properties of three materials is followed by the brief presentation of types of composite materials and their constituents. The second part is focused on the introduction and features of bolted joints. The particular interest is dedicated to the joints with composite clamped members, their solicitation and failure modes, followed by the challenges related to the preload application. The objective of this Chapter is to analyse the literature in order to foresee non-examined environmental conditions, what changes they may cause to materials and how such behaviour may impact the bolted joints.

Chapter II is focused on the experimental characterisation of given thermoplastic composites and neat matrix. Prior to mechanical characterisation, a conditioning protocol is elaborated and validated for the desiccation and humid ageing of the materials at different humidity levels. The evaluation of glass transition temperatures is accomplished in order to rigorously analyse the following experimentally obtained mechanical properties. They are studied at several temperatures and relative humidity levels that are selected individually for each material according to their in-service conditions. This campaign provides not only the rigorous in-plane elastic material characteristics but also the input and comparative data for the numerical simulations in Chapter III.

Identification of numerical elastic out-of-plane properties of the composite PA6 GF is the principal aim of Chapter III. The double-scale homogenisation approach with the use of the finite element method is proposed for the numerical identification of the mechanical parameters and their validation by comparing to the experimental data. The complete reconstruction of tow geometry and the geometry of the Representative Volume Element is presented. The necessity of this campaign is justified by the difficulties of the experimental evaluation of the out-of-plane elastic modulus and its vital importance for the study of the mechanical behaviour of bolted joints, demonstrated in Chapter IV. Furthermore, the obtained properties complete the evaluation of the whole range of composite mechanical properties from Chapter II. Eventually, these results are aimed to be integrated into the software CETIM-Cobra in order to manage the sensitivity of the composite material to given environmental conditions.

The last Chapter IV is focused on the characterisation of the behaviour of bolted composite joints subjected to preload. The compliance of composite material PA6 GF, used as the clamped parts, is initially analysed with the application of out-of-plane modulus, computed in Chapter III. This coefficient is important for preload conservation over time. Preload characterisation, demonstrated in the second section, is followed by the preparation stage. It consists of the bolt calibration and determination of preload and bolt compliance. Eventually, the results for unconditioned and conditioned composite specimens are presented and analysed. The

bolt elongation and loss of preload as a function of environmental conditions are important for the conception of bolted joints; thus, are expected to be integrated into the mentioned software.

The report finishes with Chapter, dedicated to the general review of the performed research work and proposed future perspectives. The suggested prospects provide some ideas, not implemented in the present work, to enlarge the database of material properties and to characterise bolted composite joints in more detail for improving structure design.

CHAPTER I

LITERATURE REVIEW

The present research work is focused on the durability of preloaded bolted joints with thermoplastic composite members under the impact of environmental conditions. Two composite materials and a matrix are provided for the study. By cause of material sensitivity to temperature and humidity, a review of available and relevant information regarding the environmental effects on the given materials is necessary prior to the numerical and experimental campaign. A significant sensitivity to both temperature and humid impact, resulting in changing mechanical features and glass transition temperature, is demonstrated for one composite material and matrix. Although another composite material manifests itself as mainly temperature-dependent, some small variations of properties are observed for high moisture uptakes. Hence, the first section helps to evaluate the global challenges for the material response under changing environment. The second section is dedicated to the behaviour of bolted joints with applied pretightening and composite members. A brief review of the connection types is followed by the characterisation of out-of-plane behaviour of such assemblies. The importance of preload is particularly highlighted for the applications of metallic and composite materials. The last section outlines the main interests and the axes for the present research work. Thus, the analysed physical, mechanical and chemical properties of given materials serve for the definition and elaboration of a methodology for their experimental characterisation (Chapter II). The evaluation of challenges concerning the preloaded bolted joints with clamped composite parts contributes to the numerical (Chapters III and IV) and experimental (Chapter IV) examination of the mechanical response of assemblies and their members.

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I.1. INTRODUCTION TO ENVIRONMENTAL IMPACT ON PROPER-TIES OF COMPOSITE MATERIALS

The present section details the physical and mechanical properties of two composite materials, glass-fibre reinforced polyamide 6 and carbon-fibre reinforced polyphenylene sulphide, and a matrix polyamide 6. Firstly, the introduction to the relative humidity and its importance for industrial applications and composite materials is presented. Secondly, the effects of acting temperature coupled with humidity are analysed for three study materials through the literature review. The analysed information is outlined in the last section; the scope and objectives are likewise presented.

I.1.1. Importance of humidity for industrial applications

An invisible form of water – water vapour – mainly comes from the oceans through the evaporation process. It can be also represented in the form of relative humidity in order to evaluate the humid or dry atmosphere. Hence, humidity is an essential factor for material processing, storage, testing and applications. Although the relative humidity cannot be a parameter that would characterise a material itself; the moisture content indicates its hydrophilic/hydrophobic behaviour. Resulting in the dimensional and mechanical changes, the moisture content, strongly related to the absolute humidity, is of particular importance for structure, part and joint design.

Humidity, or absolute humidity, stands for the amount of moisture in the air and is represented by the amount of water vapour per volume of air. It means that the higher the quantity of water in a given air volume, the higher the absolute humidity. What is more, it is not dependent on temperature. On the contrary, more commonly used **R**elative **H**umidity (RH) measures the amount of water vapour in relation to the air temperature **[2,3]**:

$$RH = \frac{C}{C_{max}} \cdot 100 \tag{1}$$

where C is the current water vapour content (g) and C_{max} is the maximum water vapour capacity of the air (g) at a given temperature. Thus, RH is a measure of the air saturation rate at a current temperature. Another definition of the relative humidity is the ratio of the partial vapour pressure to the saturated vapour pressure at a given temperature [2,3].



Figure I-1: Maximum absolute humidity as a function of air temperature¹

The temperature at which the saturation occurs, so when the condensation begins, is called a dew point [3]. It is an essential indicator of the amount of water vapour in the air along with the relative humidity. The air capacity to saturation and the vapour pressure are temperature-dependant, and the warm air is known for the higher capacity than the cold [3]. Nevertheless, the same amount of absolute humidity would signify that the

¹ <u>http://wps.prenhall.com/ca_ph_christopherson_phygeo_1/42/10953/2804088.cw/content/index.html</u>

cold air is of higher RH rather than the warm. This trend is demonstrated in Figure I-1: the air can contain 15 g of water vapour at +20°C, but 47 g at +40°C. Besides, the RH, along with the temperature, has a tendency to fluctuate over a day, a season and a year, as shown in Figure I-2 and Figure I-3.



The relation between moisture content and relative humidity can be established through an experimental campaign. An example of the equilibrium moisture content of wood as a function of RH is demonstrated in Figure I-4. One can notice that at one given temperature, for instance, at $+30^{\circ}$ C, the moisture content varies from roughly 2.5% to 20%. Nevertheless, this gap tends to decrease with the temperature rise. The testing temperature is of greater importance for high levels of relative humidity as in the case of 70% and 90%. Generally, material sorption capacity can be represented with a sorption isotherm (Figure I-5), a curve relating the maximum absorption capacity to the relative humidity of the surrounding atmosphere at one given temperature [4].

Even though the temperature is one of the critical impact parameters on the moisture content in materials, the RH level plays a more significant role.



Figure I-4: Moisture content of wood as a function of relative humidity and temperature **[3]**



The absorbed humidity may also provoke the dimensional changes. The material heterogeneous structure can lead to dissimilar longitudinal, transversal and out-of-plane deformations – shrinkage or expansion [3,5,6]. Such behaviour is visible in the case of wood, demonstrated by [3,6]. Its tangential and radial deformations are different but linearly related to the equilibrium moisture content. Although it may take months or years to achieve complete saturation, these varying parameters are to be taken into consideration while designing and manufacturing. Proper storage of materials before production is equally necessary in order not to change or

¹ https://www.weatheronline.co.uk

² <u>https://www.weatheronline.co.uk</u>

degrade the structure. Hence, humidity and temperature are to be regulated [7]. However, the external environmental conditions fluctuate throughout a year [8], so the moisture content does [9], and the similar but reproduced conditions in a climatic chamber may not indicate the same results [10,11].

Though the relative humidity does not illustrate the vapour content directly as the absolute humidity, it remains a widely used standard parameter to describe environmental conditions and, along with moisture content, is employed in the current work.

I.1.2. Brief introduction to composite materials

Several generalities on composite materials are presented in the current section. Some types of employed matrices, reinforcement and their applications are briefly discussed. However, in terms of the mechanical and physical properties, the main focus will be on the selected study materials, which are presented in more details in the sections I.1.3, I.1.4 and I.1.5.



Figure I-6: Types of matrices employed in composite materials **[12–14]**. The capital letters stand for abbreviations. The list is not exhaustive. Red contour stands for the materials used in the current work

A composite material, as indicates the term, is a composition of two or more constituents that do not dissolve or merge into each other. Consequently, such material possesses a heterogeneous structure, like wood, mud bricks, paper laminate, bones and teeth, concrete that are known as natural composite materials. Two main components are the reinforcement and the matrix, so the latter serves for the material classification: Metallic-Matrix Composites (MMC), Ceramic-Matrix Composites (CMC) or Polymer-Matrix Composites (PMC) (Figure I-6). Metallic and ceramic matrices are generally used in MMC and CMC materials. Polymer-based ones have two subcategories – ThermoPlastic (TP) and ThermoSet (TS) that represent nearly 90% of all composite materials. The functions of matrices are to transfer the load to reinforcement, ensure the permanence of the reinforcement geometry and protect it from the environmental impact. Nevertheless, its additional functions may be expected according to design and application requirements.

As for a reinforcing material, there exists a wide range of particles and fibres starting from the nanosize (for instance, carbon black **[15]**), short fibres to massive diameters of metallic wires (Figure I-7). The reinforcement type is selected to fulfil the requirements of an application field, compatibility with a matrix, cost etc. Its primary function is to define the mechanical properties of a composite part.

Concerning the reinforcement architecture, it is known for being either non-structured (a mat) or structured: woven, non-crimp, braided and knitted **[16–19]**. The woven reinforcement, in its turn, can be of twill, satin or plain weave, which defines the rigidity of the reinforcement **[1,18]**. This type of fibrous material is most common in composite materials due to such advantages over other structures as the moderate production cost, relatively simple technological process and possibility to obtain quasi-balanced mechanical properties.



Figure I-7: Type of reinforcement employed in composite materials **[12–14]**. The list is not exhaustive. Red contour stands for the materials utilised in the current work



Resulting from the extensive application of composite materials in various domains, the interest is currently being shifted from the thermoset to thermoplastic composites mainly due to the lower final production cost including a possibility to recycling. What is more, high chemical and impact resistance, better fatigue resistance represent other advantages of TP matrices [20–23] (Table I-1). However, some disadvantages of TP in comparison to TS matrices shall be taken into consideration. Among them, we can cite: high fabrication cost of CM with high-performance matrices, for instance, PEEK, PPS; difficulties in processing; the impact of moist and/or hot environment. Indeed, some TP and TS can be sensitive to temperature (PPS, PEEK) and moisture (PA6, PA66, epoxy), leading to degradation of some properties [4,15,24–27]. Nevertheless, the final properties of composite materials are defined by a proper combination of constituents so to be adapted to the needs of the working environment.



Figure I-9: C	Composite	materials	in the	e industry	[28]
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	Thermoset composites	Thermoplastic composites	
	High-temperature resistance	Infinite shelf life	
	Resistance to solvents	High delamination resistance	
	Resistance to creep	Recyclability	
	Low viscosity	Impact resistance	
Advantages	Low processing temperatures	Chemical resistance	
	Well established properties	No emissions	
	Excellent bonding with fibres	Shorter transformation cycles	
	Relatively cheap	Better fatigue resistance	
		Possibility of repair	
	Limited storage life	High viscosity	
	Difficulties for thick composite parts	High manufacturing temperatures	
Drawbacks	Non-recyclability	Prone to creep	
	Long processing time	Generally more expensive	
		Poor drapability	

Table I-1: Advantages and drawbacks of thermoset and thermoplastic composites [19, 21, 22]

The moisture adsorption capacity of a composite material principally depends on its composition – type of reinforcement and matrix. Organic, like glass and carbon, and metallic fibres are known for hydrophobic properties [12,13]; thus, the use of these fibres in parts, working in a humid environment, would lead to stable mechanical properties related to the reinforcement. Unlike with organic fibres, the natural ones possess hydrophilic properties [29–33]. The interaction with moisture leads to their swelling. This may result in different damage initiation mechanisms as fibre-matrix interface decohesion or fibre-fibre decohesion, for instance, after cyclic desorption-saturation experiment [32]. Despite the relatively low production cost, the natural fibres remain yet under thorough analysis and study by numerous researches, whereas the organic fibres found their

applications decades ago. Their fully assessed and independent of environmental conditions high mechanical properties in combination with well-defined processing protocols ensure the extensive use throughout numerous domains.

As for matrices, they play a definite role in the moisture absorption capacity of a composite with organic reinforcement [4,23]. This property is defined by chemical composition and pertains to particular TS and TP matrices. However, different liquids may have an untypical impact on absorption capacity. The authors of article [34] demonstrate the effect of humid air, distilled and saturated saltwater, diesel fuel, gasoline etc. on the evolution of moisture content till saturation in polyester glass composite that belongs to TS group. Another widely used TS matrix – epoxy – is also known for the ability to absorb moisture [15,25,35–40]. It is shown by the authors [36–38,40–42] that the temperature affects the duration to obtain the maximum moisture content, whereas the level of moisture content is dependent on the RH and insensitive to temperature. Besides, some matrices, for instance, polyester and epoxy, can be susceptible not only to water but also to other liquids [37]. Among a large variety of thermoplastic matrices, known for more ductile behaviour than the TS materials, some are more sensitive to humidity than the others. For instance, PolyEtherEtherKetone (PEEK) and PolyPhenylene Sulphide (PPS) filled with carbon fibres possess low maximum moisture uptake [26,39,43]. Though, other thermoplastic polymers, like PolyStyrene (PS) and PolyAmides (PA), undergo humidity impact [44–48].

For the present project, two thermoplastic composite materials with the different susceptibility to humidity impact, briefly demonstrated in the present section, are selected. Glass Fibre reinforced PolyAmide 6 (PA6 GF) and Carbon Fibre reinforced PolyPhenylene Sulphide (PPS CF) are employed in different domains due to remarkable mechanical and physical properties. Thus, PA6 GF composite material can serve for a reference in terms of its high hygroscopicity for the consequent comparison to the advanced PPS CF. In a nutshell, rigorous analysis of hygro-thermo-mechanical behaviour of PA6 GF and PPS CF will allow estimating the mechanical behaviour of bolted composite joints in different environmental conditions.

More detailed effect of environmental conditions on the employed materials is discussed in the following sections: temperature and moisture impact on a neat matrix PA6, composite materials PA6 GF and PPS CF are presented in the sections I.1.3, I.1.4 and I.1.5 respectively. The hygro-thermal impact is particularly detailed in section I.1.3, dedicated to the description of neat matrix properties since the matrix is mainly in charge of the composite sensitivity.

I.1.3. Hygro-thermal impact on chemical, physical and mechanical properties of neat matrix PA6



I.1.3.1. Some generalities on PA6

¹ Phase amorphe stands for Amorphous phase ; Lamelles cristallines stands for Crystalline lamellae ; Amas lamellaire stands for Lamellar cluster ; Structure sphérolitique stands for Spherulitic structure

Polyamides are engineering thermoplastic materials, widely used in the form of neat or reinforced matrices. Polyamide 6 is one of several grades of polyamides that are dependent on the polymerisation type. This polymer possesses a semi-crystalline structure, constituted of amorphous and crystalline phases with the degree of crystallisation around 30-40% that tends to increase with humid ageing **[49,50]** (Figure I-10). Such structure enables material recycling due to the ability of melting, which differs this type of matrix from the thermosetting. The degree of crystallinity characterises final mechanical properties, but tends to decrease with higher cooling rate and practically independent of the presence of fibres **[21]**, whereas the amorphous phase with disordered chains has different mechanical and physical properties from the arranged crystalline phase **[51]**. Mainly due to the amorphous phase, this material is therefore highly sensitive to environmental conditions, i.e. temperature and humidity, as demonstrated by many authors **[4,46,49,52–54]**.

I.1.3.2. Moisture impact

Polyamide 6 intensively absorbs water due to the presence of amide groups -HN- in the amorphous part of the polymer (Figure I-11 – Figure I-12), which has effects similar to temperature **[4,21,55]**. At first, water molecules occupy the free space like pores or cavities and then interact with hydrogen bonds, finally creating water molecules aggregate **[47]**. A certain quantity of accumulated molecules or clusters induces the activation of chain mobility and decreases the attraction between molecules, leading to the flexibility of the polymer. This interaction explains the material sensitivity resulting in plasticisation and physical degradation when in service **[15,55,56]**. The second type of degradation is chemical and irreversible from the mechanical point of view, resulting from the hydrolysis process **[5,57]**. The determined molar weight and distributions by Le Gac et al. **[58]** in unaged, aged in seawater for six months and also dried after ageing specimens did not show relevant changes. Hence, chemical damage is not considered in the current work.



Figure I-11: Chemical structure of PA6 [47]



Figure I-12: Representation of a chemical structure of PA6. Water molecules are located in yellow circles [47]

Dimensional changes

Starkweather [59] is one of the pioneers who commenced examining nylons in moist environments. He discovered the quasi-linear relation between dimensional expansion and mass changes of polyamide 66 due to humidity (PA66 is very similar to PA6 in terms of their moisture uptake capacities). It is, however, valid on a large scale, and, as demonstrated by [4,60,61], some non-linearities are present at the beginning of the water sorption (Figure I-13). Such behaviour can be explained by an initial occupation of a present free volume that does not contribute to noticeable expansion. It is worth mentioning that the water molecules are not homogeneously distributed throughout a material thickness and length, hence, creating water profiles [25]. Nevertheless, the longitudinal and out-of-plane deformations are coherent to the present water content [62–65]. Such time-dependent water profile is affected by a diffusion coefficient, proposed in [66], and boundary conditions, whereas the time to desiccation/saturation is also determined by material geometry. The complexity of specimen geometry on the water uptake is investigated and demonstrated by [67–70]. They assign it to the



non-linear relation of the specimen thickness and time to saturation (Figure I-14). Also, the authors clearly show that a bigger thickness leads to a significant increase in time, needed to obtain a required material condition.

Figure I-13: Deformation ε_{xx} (%) of neat matrix coupons of PA6 as a function of a global water content C_{glob} (%) [4]



Figure I-14: Moisture content versus square root of time [68]

Moisture desorption

Material tendency to moisture desorption and its amount are characterised by several parameters (Figure I-15 – Figure I-16). First of all, the final loss of mass is dependent on the sensitivity of the material to humidity and the initial moisture content. The desorption kinetics are affected by the type of drying environment. For instance, high temperatures induce more rapid moisture losses that can be enhanced by made vacuum. However, as already mentioned, prolonged exposure to extreme temperatures may result in chemical degradation with the consequent mass loss, which is no longer related to moisture loss, but structure degradation. What is also interesting to mention is that the sanded and normal coupons demonstrate quasi-identical results during desorption as shown by Obeid [4], so that the damaged surface of material does not lead to faster or slower material desorption capacity.

Theoretically, in **D**ry-**A**s-**M**oulded state (DAM) material should not contain any moisture. Nevertheless, in real conditions, the desorption process does not lead to complete material desiccation. Consequently, insignificant moisture content of 0.2% can be present, as demonstrated in **[69]**.





Figure I-15: Moisture losses of neat PA6 coupons employing different methods. Abscissa axis stands for the square root of time. Test duration is 50 days [4]

Figure I-16: Water desorption versus square root of time in the 4.1-mm-thick samples [46]

Moisture absorption

The effect of RH on the absorbed amount of moisture by PA6 and its kinetics was studied by [4, 5, 21, 49, 67]. The authors show that the linearity only exists until an approximate threshold of RH 85%. Beyond this

value, the slope increases, illustrating a more important uptake capacity (Figure I-17). Krzyzak et al., Arboleda-Clemente et al. **[54, 69]** examined the moisture uptake by PA6. At the same conditioning temperature +23°C, the PA6 absorbs 3% of moisture at RH 50%, 3.5% at RH 60% and 9.5% when saturated in water. Silva et al., Starkova et al., Taktak et al. **[52, 71, 72]** explored the effect of temperatures from +20°C to +90°C on the absorption and desorption kinetics and showed that the saturation and the complete desiccation occur the fastest at +90°C. Arhant **[21]** also demonstrates the influence of high temperature on the water uptake (Figure I-18). However, the amounts of absorbed and then consequently released water at the same temperature, are not equal. The authors relate this phenomenon to the bound water of the Langmuir model.



The water diffusion within PA6 matrix is frequently modelled with a common Fickian law [73] that represents the diffusion behaviour of many materials. It is adopted by numerous researchers due to its simplicity and straightforward experimental identification: at the beginning of the sorption, the curve shall be linear with the consequent asymptotic value [4, 21, 47]. The theoretical Fickian law is shown in Figure I-19 with the dashed line. The experimental line, however, reveals some differences from the widely used law [53, 74]. Arhant [53] developed a model that describes the changing slope more precisely, as illustrated in Figure I-20. Nevertheless, with increasing experimental temperature, the dissimilarity of Fickian and experimental data tends to reduce as shown in [21] for immersion at +40°C and +60°C.





Figure I-19: Water content of PA6 immersed in seawater at +15°C: comparison between experimental results and Fickian law [53]

Figure I-20: Comparison between a developed model, experimental results and Fickian model of immersed into water at +25°C thick specimen **[53]**

Glass transition temperature

The nature of the amorphous phase is such that its chains have a possibility to move. However, the movement amplitude is determined by two regimes, delimited by a glass transition temperature T_q (Figure I-21 –

Figure I-22). Below it the material is in glassy state, hence, the amorphous chains do not deform due to the thermal agitation insufficiency and both phases, crystalline and amorphous, are fragile. Above T_g , the amorphous phase is in viscous (rubbery) state due to sufficient thermal agitation, leading to large deformations [47]. Consequently, T_g lowers robustness and rigidity by increasing energy absorption and ductility of polyamides [69].



The glass transition temperature is the subject of numerous articles. It can be estimated experimentally with **D**ynamic Mechanical Analysis (DMA) or **D**ifferential Scanning Calorimeter (DSC) (Figure I-22). Besides, T_g of polyamides is highly dependent on the content of absorbed water. For instance, [76] analysed its dependence on the moisture content of oriented polycaproamide. A drop in the T_g is nearly +70°C: from +60°C for a dry material to around -10°C when containing approximately 9% of moisture at saturation (Figure I-23 – Figure I-24). Same results were likewise obtained by [23, 54, 58, 62, 77, 78] for the other polyamides, including PA6. Similarly, Le Gac et al. [58], Ishisaka et al. [55] 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.





Figure I-23: Glass transition temperature of PA6: experimental data compared to theoretical [21]

Figure I-24: Glass transition temperature of PA6 for the moisture content between 0% and 10.3%, measured by DMA with the shear method **[79]**

Mechanical properties

Tensile and compressive properties

The moisture uptake by a neat polyamide induces the changes in mechanical behaviour, resulting in a smoother stress-strain curve **[80]** (Figure I-25). However, the definition of an elastic limit becomes more complex in this case.



Figure I-25: Mechanical response of polyamide in dry state and with moisture content [80]

Relevant studies towards the understanding of the water-induced mechanical changes of neat and reinforced PA6, or similar PA66, were carried out by **[23,45–47,52,58,67,77,78,81]**. According to Taktak et al. **[52]**, Silva et al. **[71]**, Le Gac et al. **[58]**, tensile Young's modulus, the yield stress and the ultimate stress tend to a remarkable decrease of nearly 80-90% when the water content is about 9% (Figure I-26). Hence, the linear elastic region is clearly visible to a threshold of water content, for instance, up to 1.7%, but above this limit, the plastic region is much pronounced. The analysis of the in-plane tensile extension, demonstrated by **[23,45,52,67,81]**, show the non-linear relation of a tensile strain at rupture and moisture content, or RH. The elongation is assumed to increase with water content till an admissible level of the latter, after which the strain tends to reduce. The compressive stress is analogously related to the moisture uptake, inducing higher amounts of strain and lower compressive resistance as depicted in Figure I-27 and Table I-2 **[80]**.





Figure I-26: Cyclic true stress-strain curves of PA66 at +23°C for different RH in quasi-static loading [23]

Figure I-27: Influence of water absorption on the compressive stress of non-filled PA6 at 25°C, the deformation rate is 10%/min [80]

Category	Unit	Nylon 6 CM1017	Nylon 610 CM2001	Nylon 66 CM3001-N
Yield stress (in dry state)	MPa	85	90	65
Yield strain (in dry state)	%	5	5	5.5
Stress at 1% of compressive deformation				
In dry state	MPa	26	20	28
At equilibrium water absorption	MPa	5.5 (3.8)	7.2 (2.2)	8.0 (3.7)
At saturation water absorption	MPa	3.4 (11.5)	5.2 (3.8)	4.9 (9.9)

Table I-2: Compressive strength. Numbers in parentheses represent water absorption in % [80]

Creep

Creep resistance is one of the most important mechanical properties of materials. Polymer materials can deform while subjected to the stresses below the 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 long-time exposure to mechanical stresses. Hence, under long-term exposure to applied stresses, the material deformation can reach large magnitude, so the part may no longer fulfil its function. Polyamide 6 manifests itself as a viscoelastic material under the influence of temperature (see I.1.3.3).

The resistance of the material to creep over time is also degraded with the absorbed moisture **[4,47]** (Figure I-28), and the presence of water intensifies the viscous deformation of PA6. What is more, the minimum velocity of creep deformation under traction evolves linearly and is higher as compared to the unaged matrix, which possesses two regimes of deformation rate **[47]** (Figure I-29).



I.1.3.3. Thermal impact

Not only the presence of water molecules entails the degradation of mechanical properties, but also the temperature, as stated by [82]. It is attributed to increased chains mobility above T_g . Prolonged exposure to elevated temperatures results not only in the deterioration of physical, but also chemical properties due to thermo-oxidative ageing. The molar mass of the polymer drops drastically from the beginning of exposure (Figure I-30 – Figure I-31) due to a process occurring prior to chain cleavage [5]. It is obvious from both figures that the velocity of chain cleavage is strongly related to the temperature and tends to increase with its rise.



Mechanical properties

Resulting from thermal exposure, all the mechanical properties are affected. Due to the particular interest to some solicitation modes in the research work, we will only focus on tensile, compressive and creep behaviour.

Tensile and compression properties

Tensile strength of PA66, similar to PA6, was investigated by Bernstein et al. [57] by exposing the neat matrix to the thermo-oxidative and hydrolytic ageing in oxygen. Authors demonstrate the reduced mechanical performance over time at both high and low temperatures (Figure I-32). Their study allows predicting the remaining tensile strength of the matrix at +21°C and +37°C for several decades. Tensile modulus is therefore sensitive as well and is associated with T_g being around +50°C – +60°C in the case demonstrated in Figure I-33.



Similarly to tension, the compressive stress of neat PA6 significantly varies with the influence of experimental temperature (Table I-2). Likewise for the moisture ageing, the increasing temperature from -40°C to +100°C the compressive stress tends to decrease in a non-linear manner as presented in Figure I-34 [80].



Figure I-34: Influence of the temperature on the compressive stress of non-filled PA6: dry state, the strain rate is 10%/min [80]

Creep properties

Numerous researches dedicated their works to the analysis of creep behaviour of polymers and polyamides in particular. For instance, Yang et al. [83] showed that the creep deformation of PA66 filled with nanoparticles was higher at +50°C than at +23°C, which is related to T_g being close to +50°C. Hence, the exposure to +80°C lowered the load-bearing capacity of specimens and increased the extension of PA66 (Figure I-35). Chevali et al. [84] reported very low creep compliance of PA66 during flexural creep tests, which slightly increased from +23°C to +90°C. The creep rate of PA6 is strongly dependent on testing temperature and stress level as demonstrated by Lyons [82] for +23°C, +100°C and +150°C. This tendency to increasing creep deformation is also mentioned by [75,80,85] (Figure I-36).



nanocomposites tested under 40 MPa and (a) 23°C, (b) 50°C and (c) 80°C [83]

Figure I-36: Tensile creep deformation at 20 MPa of CM1017 - PA6 in dry state at different temperatures [80]

I.1.3.4. Summary

The hygro-thermal environment is demonstrated to have a significant influence on a wide range of neat matrix properties. The moisture is absorbed by PA6 due to the amide groups that partially compose the amorphous part. Created clusters from the interaction with hydrogen bonds activate the chain mobility, which eventually leads to physical degradation and polymer plasticisation. Absorbed water results in mass change and material expansion. However, this matrix possesses reversible properties; thus, it returns to the initial state after desiccation. The kinetics of absorption and desorption are not identical and, in addition, are impacted by the specimen geometry, in particular, thickness. The glass transition temperature is likewise affected with a strong tendency to decrease with the higher moisture uptake. Besides, the working temperature and surrounding humidity form a coupled condition resulting in changing mechanical behaviour. As a result, a thorough material

analysis requires not only a wide spectrum of testing conditions, including extreme in-service temperatures and moisture contents but also different specimen geometries.

I.1.4. Hygro-thermal impact on chemical, physical and mechanical properties of composite PA6 GF

I.1.4.1. Some generalities on PA6 GF

Polyamide-based composite materials are employed in numerous parts for the subsequent implementations in the automobile industry, for instance, clutch pedals, clutch master cylinders, steering wheel, levers, seat frames, cooling fans, rocker box covers, air filter nozzle etc. **[4,86]**, so they endure an important hydrothermal mechanical impact. These components are required to sustain a wide range of temperatures, from -40°C to +80°C, and dry/humid atmospheres in order to fulfil their functions. Such extensive employment requires an indepth and detailed analysis; consequently, this composite material has been a subject of numerous studies and articles for the recent years, summarised in the next sections.

The PA6 GF is a composite material with the thermoplastic matrix polyamide 6. As mentioned in the section I.1.2, fibres, except organic, are presumed not to undergo important changes at high/cold temperatures and humid climate **[74,87]**, whereas a matrix degradation can be crucial up to the loss of its primary functions – fibre bonding and protecting. Therefore, prolonged exposure of a composite to harsh environments provokes physical and mechanical deterioration of composite materials in a severe way (section I.1.3 for the matrix properties) **[88–90]**. It is the case with PA6 GF composite as described hereafter.

I.1.4.2. Moisture impact

The presence of polyamide 6 as a constituent enables the composite material to absorb moisture throughout the service life, leading to higher ductility due to matrix plasticisation. Water is not absorbed by glass fibres (section I.1.2); hence, the penetration cannot be straightforward. Fibres, acting as a barrier, limit the movement of water molecules obliging them to deflect in some cases as demonstrated in Figure I-37. For the case of a woven structure, the penetration along the length or the width of coupons can be direct (on the left in Figure I-37). The through-thickness travel path is, however, not straightforward and can be graphically depicted as in Figure I-37 on the right.

Dhakal et al. [30], Espert et al. [91], Karmaker [92] propose three mechanisms of moisture diffusion within a composite material. The first mechanism represents the location of water molecules into cavities, cracks and other flaws on the matrix-fibre interfaces and in the matrix itself if imperfections are present. This may, however, lead to the weaker stress transfer between fibres due to the interface degradation. The second is related to the penetration of molecules inside the polymer chains of the amorphous part of the matrix and reaction with -NH-groups (section I.1.3.2). And third suggested mechanism consists of the transportation of micro-cracks appearing due to the fibre swelling, but mainly concerning natural fibres. Another water absorption mechanism can also take place if the material is exposed to mechanical loading. In that case, water molecules may occupy the newly-formed cracks and cavities due to traction, shear, bending etc. This would result in increased or more rapid water uptake during experiments [93,94].



Figure I-37: Illustration of a travel path of water molecules: along fibre orientation on the left; perpendicular to fibres or through the coupon thickness on the right **[25]**

Moisture desorption and absorption

Moisture uptake is principally driven by the matrix and its content within the composite. Resulting from this, the hygroscopic behaviour of the composite, based on polyamide 6, follows the same tendency as, for instance, Fickian law [21]. The duration to mass stabilisation is, however, different from the matrix for both desorption and absorption. The maximum admissible moisture content during desiccation is also different from the maximum capacity of the neat matrix due to the presence of reinforcement. For instance, the content of moisture desorption (Figure I-38) of the 45°-oriented specimen of PA66 GF is around 0.2 %, whereas the neat matrix losses 4 times more [4].





Figure I-38: Desorption kinetics of glass/PA66 as a function of the square root of time divided by specimen thickness of woven structure with fibres at 45° [4]

Figure I-39: Water content in carbon/PA6 composite as a result of exposure to the humid environment and immersion into water [21]

Identically to desorption, the composite absorption capacity is restricted and dependent on the fibre content. PA6 CF specimens, immersed in water, absorb around 3 %, whereas the neat PA6 matrix takes in already 10 % (Figure I-18) [21]. As in the case with the matrix, the absorption kinetics of the composite is strongly dependent on RH, environmental temperature and specimen thickness [64,95].

Glass transition temperature

Initial neat matrix behaviour may be different from the matrix itself due to processing at high temperatures in order to form a composite. Some composite material properties are therefore matrix-driven, and this is the case of the glass transition temperature. For example, the neat PA6 resin and the PA6 matrix possess similar behaviour, resulting from their T_g correspondence as demonstrated in Figure I-40 by Arhant in his thesis [21] with an only significant difference at high water activity. As in the case with the neat PA6 (section I.1.3.2), T_g decreases from +66°C when in dry condition to -12°C when fully saturated in water (Figure I-40 – Figure I-41). These results are in agreement with [96] for PA66 GF 35%.



Figure I-40: Glass transition temperature of a composite and PA6: experimental results and prediction [21]

Figure I-41: Glass transition temperature of carbon/PA6 composite saturated at different humidity conditions [21]

Damage





Figure I-42: Transverse cracking of the matrix (a) and decohesion at the interface fibre-matrix (b) in UD composite U.D AS4/3502 [99]



Resulting from the moisture diffusion mechanisms, the naturally formed flaws in the matrix become filled with water molecules during absorption. Along with the surrounding swallowing chains, this leads to the formation of damage sites inside the matrix. Pores and cavities at the fibre-matrix interface are similarly occupied, causing internal stress by reason of matrix swelling. And it may also lower the stress transfer from the matrix to the reinforcement. Due to the weak interfacial linking forces, thus, the degraded interface, such mechanisms can provoke a fibre debonding as also demonstrated by Ray [97] (Figure I-42 b); or, even, a separation of a bundle of fibres generating the crack propagation as in the case of a UniDirectional (UD) material (Figure I-42 a) [97–99]. Likewise, Chen et al. [100] mentioned a significant decrease of InterFacial Shear Strength (IFSS) in bamboo/vinyl ester composite due to humid impact (Figure I-43) on the interfacial bonding, since two other elements as fibres and matrix did not contribute to the change of IFSS.



Figure I-44: PA66 GF 30%, RH 0% specimen, (a) damage at fibre end and fibre breakages at 33% of flexural stress, (b) damage at fibre end and high plastic deformation between two adjacent fibres at 48% of flexural stress, and (c) damage propagation along fibre-matrix interface at 85% of flexural stress [101].

The in-site Scanning Electron Microscope (SEM) bending tests allowed Arif et al. [101] to reveal the weakest sites in PA66 GF 30% composite material in dry and humid states (Figure I-44). According to the authors, the damage initiates at fibre end and fibre breakage appears at low loading, then the plastic deformation occurs between two adjacent fibres leading to the consequent damage propagation along the fibre-matrix interface. Similar results are achieved by [102]. However, with the moisture uptake, the damage initiation mechanisms change; thus, at RH 50% and 30 % of flexural stress the interfacial debonding represents the damage initiation. It is followed by the crack propagation along the fibre-matrix interface in the in-plane and out-of-plane directions that is in agreement with [87]. The damage initiation at RH 100% is similar and also starts at 30% of flexural stress [101]. Consequently, we can conclude that the damage initiation and propagation sites are different for the

dry composite and the humid-aged one; and, in addition, they are of more complex nature with the presence of humidity. However, the presence of damage can be detected visually after prolonged exposure to external harsh environments, as, for instance, shown by **[8,10,11]**.

Changes in mechanical properties

Tensile and compression

Likewise to the resin PA6 (section I.1.3), water and humidity are known to have an important impact on the mechanical properties on the composite PA6 GF and, therefore, its matrix. Arising from the altering glass transition temperature **[4,23,54,69,103]** due to matrix plasticisation, the mechanical behaviour undergoes significant changes as compared to its dry state. An example of stress-strain response of a PA6 GF 30% is illustrated in Figure I-45 and the tensile strength as a function of moisture content is in Figure I-46. Although the hygroscopic impact is coupled with the thermal, we can globally notice the variation from glassy to ductile state at the negative temperature. It is, however, valid for the matrix-dominated parameters, as, for instance, shear or creep properties. In the case when they are entirely fibre-dependent, such difference does not occur as mentioned by Dau and Arhant in their thesis **[21,23]**. Nevertheless, these changes have the physical nature and, thus, are reversible to some extent **[15,56]**.

Hygroscopic swelling may initiate local internal stresses through the material thickness. This aspect was analysed by [4,25,63,64,94,104–106]. We can notice from Figure I-47 that the out-of-plane stress σ_{zz} tends to stabilise through the coupon thickness over a large period of time. In addition, as described in the previous section, the absorbed water molecules provoke the degradation of the fibre-matrix interface, which, by consequence, results in the vulnerable mechanical properties and more rapid damage [100,101, 103].



Figure I-45: Influence of temperature on the tensile stress of conditioned PA6 GF **[86]**



Figure I-46: Impact of temperature and humidity on the tensile strength of PA6 GF [86]



Figure I-47: Examples of stress profiles σ_{zz} in UD laminate of 90° of a half thickness during humid ageing; the path (a) passes through the fibres and (b) that does not pass through the fibres [4]

Bending



Figure I-48: Flexural stress - strain curves versus glass fibres weight fractions [90]

Flexural behaviour of desiccated and water-aged PA66 GF specimens is investigated by Autay et al. **[90]** and illustrated in Figure I-48. As a result of drying, the composite material demonstrates the growth of flexural strength and flexural modulus that is visible from the slope inclination. However, the specimens, immersed in water at +100°C for 8 h 30 min for hygro-thermal ageing, confirm the unfavourable effect of such conditions. They lead to the drop in flexural strength for around 40%, and since the values of flexural strain remain similar, it shows a significant decrease in flexural modulus as compared to the dry materials states. It is interesting to mention that increasing fibre content, from 0% to 30%, ameliorates the mechanical response of given materials **[90,102]**.

I.1.4.3. Thermal impact

Likewise to the neat PA6 (section I.1.3.3), the thermal effect on the composite material is associated with the glass transition temperature. If exceeded, the material state changes from rigid to viscous and one can see a drop in mechanical strengths and elastic moduli. Nevertheless, it occurs when a composite is loaded in matrix-dominated directions.

The thermal action is important not only when the material is in service, but also during the last manufacturing stages. For instance, the cooling rate has an impact on the degree of crystallinity, which is introduced in section I.1.3.1. Consequently, the mechanic properties change: high degree of crystallinity results in high modulus. It can be also explained by the reduction of the amorphous part of the material. And, as demonstrated by Arhant [21] for carbon-fibre reinforced PA6 (PA6 CF), a wide range of cooling rates is required in order to clearly see the changes in mechanical properties, even though the changes in the degree of crystallinity are visible.

As analysed by Ray [107], the thermal conditioning of an in-service composite can lead to residual stresses due to unequal thermal expansion between fibres and matrix, resulting in the debonding at the interface. This damage mechanism can be intensified if a sudden cooling occurs, from a positive to a negative temperature
Mechanical properties

Tension

Esmaeillou **[22]** studied the temperature effect on the mechanical behaviour of the injected PA66 reinforced with 30% of glass fibres that are longitudinally oriented in the majority. As a result of tensile testing, the author mentioned a strong reduction of the tensile stresses and higher strains with the temperature increase as shown in Figure I-49 and Figure I-50. It is, therefore, more important at +75°C and +90°C which are above the glass transition temperature.



The elastic modulus and tensile strength are equally affected by thermal exposure to the range of temperatures from -40°C up to +150°C (Figure I-50) **[86]**. Young's modulus has decreased by about 60% at +150°C in comparison to -40°C. Regarding the material strength, it has reduced by about 70% at the same conditions.



Figure I-50: Effect of temperature on tensile behaviour of dry-asmoulded PA6 GF 33% [86]



Figure I-51: Effect of temperature on tensile strength and Young's modulus of dry-as-moulded PA6 GF 33% [86]

Creep

The bending and tensile creep behaviour of injection moulded PA66 GF was partially studied by Hadid et al. **[108]**, Chevali et al. **[84]** and Lyons **[82]** (Figure I-52 – Figure I-53). The presented results of creep tests demonstrate a strong relation to the applied stress levels and thermal loading. We can expect the rise of creep strains at the temperatures, surpassing the glass transition **[82]**. The composite possesses lower creep compliance in comparison to the neat matrix; hence, the authors conclude that the restriction of matrix movement by fibres takes place along with the reduced deformations.





Figure I-53: Temperature-compliance data obtained over the six systems used in this study. Creep compliance values were chosen at four testing temperatures in the service temperature range of the polymers. All readings are taken at a constant time value of 30000 s **[84]**

Globally, the demonstrated composite material behaviour is very similar to that of the matrix if the charge is mainly taken by matrix.

I.1.4.4. Summary

The glass-fibre reinforced polyamide 6 undergoes various temperatures, from negative to positive, and dry/humid environment due to its application domains. However, this composite material inherits its sensitivity to the hygro-thermal environment from its matrix PA6, analysed in the previous section I.1.3. Such impact may lead to a crucial degradation of properties due to the matrix in particular since fibres do not absorb moisture. Nevertheless, they act as a barrier that forces water molecules to deviate or limits their movement inside the structure. Similarly to PA6, the composite glass transition temperature goes through the same changes by the reason that it is governed by the matrix. Hence, matrix-dominated properties as, for instance, shear, become changed. This variation is of physical nature and is reversible to a certain extent. Regarding fibre-dominated behaviour, no significant modifications are revealed. Furthermore, mechanical properties are reduced under thermal impact. Hence, the impact of acting together temperature and humidity is greatly intensified, possibly resulting in a decreased creep resistance of the material, which is of absolute importance for bolted joints. The presented literature review facilitates the selection of testing temperatures and levels of relative humidity for the experimental campaign. Besides, a proposition to establish a conditioning method is grounded on the absence of such in analysed literature.

I.1.5. Hygro-thermal impact on chemical, physical and mechanical properties of composite PPS CF

PPS CF, a "high performance" composite, is widely used in the aviation and automobile industries due to its excellent mechanical properties at different temperatures [27,109–111], its resistance to chemicals [112], its hydrophobic properties [26,109], thermal stability [112]. The matrix PPS is also employed in the electronics industry. The brief overview of the composite structure, physical and mechanical behaviour is presented in the following sections. However, the composite material is not as widely studied as PA6 GF by a research community; hence, the known information concerning the composite and its matrix is restricted.

I.1.5.1. Generalities on PPS CF

The matrix of the composite material is a thermoplastic resin PPS of the semi-crystalline structure. It, therefore, contains a crystalline and an amorphous part as described in section I.1.3.1 for PA6 matrix. Its elementary cell and crystallised structure are demonstrated in Figure I-54. The final matrix properties are highly dependent on the degree of crystallinity, which is around 60-65%, and polymeric morphology as cited by Nohara

et al. [113], Lu et al. [114], Lou t al. [115]. It is interesting to note that during the composite manufacturing a partially melted matrix can contain crystallised regions that may cause voids and fibre breakages when pressure is applied (Figure I-55). Consequently, the processing temperature of the material is relatively high, around +340°C as used and studied by [116] and [113] respectively.





Figure I-54: PPS elementary cell consisting of aromatic rings linked by sulphide **[113]**

Figure I-55: Spherulitic structure : crystallisation of PPS by using a polarised light microscope, isotherm for 28 min at +265°C [113]

The degree of crystallinity in composites undergoes very small changes after the exposure to humid ageing at $+70^{\circ}$ C (increase by 5.1% and 6.5%), whereas the matrix does not change according to Blond et al. [109], but the exposure to $+80^{\circ}$ C and 85% RH for a long period may change the crystalline fraction [117]. Although, according to Lu et al. [114], when the crystallinity decreases, the impact strength of the matrix tends to increase.

I.1.5.2. Hygro-thermal impact

During operating conditions, composite material structures may be subjected to various environmental conditions, which are mainly temperature and humidity variations. As in the case with sensitive materials, hygro-thermal ageing can lead to the slight deterioration of different physical properties of high-performance composites, especially at extreme temperatures and in very humid environments **[111,118]**. The diffusion of water within the material generally contributes to its volume expansion and higher ductility. The water content is known to be very low for PPS CF and, therefore, often negligible for engineers and scientists.

Moisture absorption/desorption

The poor moisture uptake by PPS CF can be associated with the gas phase prevailing the water clusters in the hydrophobic composites **[4,119,120]**. Hence, the researches usually expose the PPS-based material to high temperatures for some period of time in order to obtain it in the dry state. As for the absorption, more articles are found. According to the research, conducted by Ma et al. **[26]**, the quantity of absorbed moisture is barely 0.059% at $+80^{\circ}$ C/RH 75% and 0.13% at $+80^{\circ}$ C/RH 85%, though these environmental conditions are usually considered as severe (Figure I-56 – Figure I-57). Nevertheless, the stabilisation occurs fast, within two-three days. As for the diffusion coefficient, its sensitivity to temperature is not pronounced, as shown in Table I-3.

Temperature	+60°C	+70°C	+80°C
Diffusivity $ imes$ 10^{-9} , cm^2/s	35.8	36.0	37.9

Table I-3: Diffusion coefficient of water in a composite PPS CF at various temperatures and RH 75% [26]



Figure I-56: Water absorption in PPS CF composite (fibre content 65%) as a function of immersion time at various environments: (Δ) +60°C, (O) +70°C, (\Box)+80°C, RH 75% [26]



Figure I-57: Water absorption of PPS CF as a function of reduced time at various environments: (Δ) +60°C. (O) +70°C, (□) +80°C RH 85%, (■) +80°C, annealed. PPS CF was annealed at +204°C for 2 h before exposure to moisture [**26**]



Figure I-58: Water absorption vs time in consolidated, stamped PPS CF laminates and neat PPS [109]

Blond et al. [109] examined the moisture absorption by a neat PPS and two PPS CF laminates with different manufacturing approach: stamping and consolidation (Figure I-58). Previously dried specimens are exposed to $+70^{\circ}$ C/RH 85% for 1400 h for the matrix and 2200 h for composites. The authors fitted the experimental data with the Fickian law (more details in section I.1.3.2). The consequent moisture uptake by materials is the next: 0.088% for the neat PPS, 0.074% for the stamped PPS CF and 0.076% for the consolidated PPS CF. Though minor difference is present, the order of magnitude of these values is the same as for the PPS CF with the other fibre content, demonstrated in Figure I-57 by [26,121,120].

Glass transition temperature

Cho et al. [116] investigated the glass transition temperature of a neat and filled with carbon nanofibres at different heating rates. Though only slight variations of T_g were found (Figure I-59), the crystallinity becomes lower for higher proportions of nanoparticles, which tend to act as obstacles for the proper arrangement of molecules. The drop in storage modulus is practically the same for all the compositions (Figure I-60).

The increase in T_g with the immersion time and temperature, as illustrated in Figure I-61, may indicate the thermal effect occurring in the composite [26]. One can presume that it is related to the post-curing, particularly highlighted above the initial T_g of +90°C. The examined T_g of PPS CF by [109] is, however, higher than



demonstrated by other authors [26,116,121], around +112°C – +113°C with an increase of +1°C after humid ageing.

Figure I-59: Differential scanning calorimetry (DSC) analysis of PPS-CNF composites as a function of heating rate 5 °C/min. CNF proportions are by volume [116]



Figure I-60: Dynamic mechanical thermal analysis (DMTA) of PPS-CNF composites as a function of temperature. CNF proportions are by volume **[116]**



Figure I-61: Glass transition temperature of PPS CF composite versus immersion time at various environments: (Δ) 60°C, (\Box) 80°C, (O) 95°C, RH 75 % [26]

Mechanical properties

The present thermoplastic polymer PPS is known for its thermal resistance due to high glass transition and melting temperatures of around +90°C and +285°C respectively. The neat PPS matrix is known to be rigid and brittle. It is caused by the presence of large spherulites in the structure **[27,114]**.

Tensile and compressive properties

According to [26], the temperature-dependent tensile strength of the composite material decreases at similar rates at the temperatures below T_g , but this reduction is more rapid when above T_g (Figure I-62). Nevertheless, one can see the tendency to the strength stabilisation over time to approximately the same level in spite of a used temperature. Hence, one can conclude that the mechanical behaviour of PPS CF tends to change in the vicinity of T_g . Similar results are received by [27,115] (Figure I-63). According to the authors, the tensile and compressive strengths tend to decrease remarkably when beyond T_g . What is interesting is that the compressive

and tensile properties level off after a long time exposure to water **[115]** as we can see in Figure I-63 and partially in Figure I-62.



Figure I-62: Tensile strength of PPS CF composite versus immersion time at various environments: (Δ) 60°C, (\Box) 80°C, (O) 95°C, RH 75 % [26]



Figure I-63: Effects of hot water exposure on strength retention of PPS reinforced with long continuous glass fibres [115]

According to Figure I-64, the stress-strain response is independent of such environmental conditions as humid ageing and temperature as studied by Vieille et al. **[43,111,118,122]**. This elastic behaviour is predictable by reason of fibre domination in the case of quasi-isotropic laminates.

The applied compression to PPS CF laminate results in the typical stress-strain curves, demonstrated in Figure I-65. According to Vieille et al. **[110]**, the compressive strength and modulus tend to decrease by 30% and 9% respectively under severe conditions.



Figure I-64: Influence of severe conditions on unnotched quasiisotropic laminate PPS CF [43]



Figure I-65: Influence of stamping on the compressive behaviour of [(0/90)]₇ PPS CF laminates under severe conditions: loaddisplacement responses **[110]**

Flexural properties

When it comes to the flexural strength (Figure I-66 – Figure I-67), the temperature impact is reversed as compared to Figure I-62: the reduction at +95°C is slower than at the temperatures below T_g ; however, the stabilisation occurs over time with similar final values [26]. Globally, as in the case with the tensile properties, the proximity to T_g induces the changes in bending behaviour that are in accordance with the results from [27].

Flexural behaviour is analysed in more details by Ma et al. [117] (Figure I-67). Composite material PPS CF, immersed in water at temperatures +70°C and +80°C for 120 days, demonstrates the retention of initial properties by about 55%-75%, which is in accordance with [115]. Hence, the prolonged immersion in water along with the thermal action may indeed deteriorate the composite material structure and, thus, its properties.



immersion time at various environments: (Δ) 60°C, (\Box) 80°C, (O)

95°C, RH 75 % [26]



As for the temperature dependence, the bending stress at rupture exhibits an important relation to such condition with the consequent decrease when the increase of temperature takes place **[123]**.

Figure I-67: Effect of water immersion on the retention of flexural strength of the PPS CF composite. *Annealing condition is 204°C for 2 h **[117]**

Shear properties

With regard to the thermo-mechanical behaviour of the off-axis woven composite, it is mainly guided by the behaviour of its matrix. For a thermal condition below T_g , the composite shows a linear stress-strain relation representing the elastic properties at the beginning of the test, which becomes non-linear from the beginning of loading due to the viscoplastic nature of the matrix [124–127] if above T_g . Consequently, in laminates with 45°-fibre orientation the response changes for an elastic-ductile with a significant drop of shear modulus and strength, and an increase of shear strain at failure (Figure I-68). The onset of plastic deformation, represented by a non-linear region between 17% and 30% of shear strain, is clearly visible at hostile conditions. Nevertheless, the composite retains sufficiently its mechanical properties after the environmental impact. De Baere et al. [127] used different shear testing methods, which, however, gave similar shear stress – shear strain curves as illustrated in Figure I-69.

Figure I-70 emphasises the ductile behaviour of the matrix PPS at the ambient temperature, which becomes intensified when exposed to +120°C. It directly contributes to the matrix-dominated shear of the composite that undergoes an increase of strain, a decrease of shear strength and modulus **[109]**.



The interlaminar shear response, obtained with a three-point bending under the hygro-thermal impact, manifests also the ductile behaviour of matrix-rich regions, which are more present in the consolidated laminate.

Nevertheless, the interlaminar shear strength gets reduced by 40% for each type of laminate in comparison to the testing at ambient temperature **[109,110]**.



Figure I-70: Influence of temperature on the tensile response of [±45]₇ PPS CF laminates (in-plane shear) and neat PPS specimens [109]



Figure I-71: Influence of stamping on the interlaminar shear behaviour of quasi-isotropic PPS CF laminates under severe conditions: load-displacement responses [110]

Damage

Resulting from the tensile testing on PPS CF in dry-as-moulded and severe states, Vieille et al. **[43]** observed the progressive rotation of fibres. It is drawn on the surface of a specimen in Figure I-72. For DAM condition, the fibre orientation angle is around 28° at the end of the test, whereas at +120°C with the prior humid ageing the angle is 19° instead of initial 45° **[43]**. What is more, the significant delamination is associated with the necking (Figure I-72 on the right). Authors suggest that the damage occurs due to the decrease of matrix strength, its shear modulus and also due to the reduction of bonded forces between matrix and fibres, thus, lower interlaminar shear stress. This reasoning is in agreement with the damage mechanisms of woven and UD composites, discussed in section I.1.4.2, and the delamination in UD PPS CF composite cited in **[115]**.



Figure I-72: Observation of angle-ply PPS CF unnotched laminates at $T > T_g$ before ultimate failure: (on the left) rotation of fibre bundles and (on the right) extensive delamination associated with the rotation of fibres [43]

I.1.5.3. Summary

The presented thermoplastic composite material PPS CF belongs to advanced materials due to its excellent mechanical properties, resistance to chemicals, temperature and humid impact. The mechanical behaviour is strongly related to the degree of crystallinity of the matrix. It is worth mentioning that the crystalline part in composite tends to increase after exposure to humid impact, whereas no changes are observed in the neat matrix PPS. However, the humid impact is generally neglected by manufacturers due to very small moisture uptake. This seems to be admissible for the applications where the properties are dominated by the fibres; however, the matrix-dependent shear and bending behaviour may be affected; hence, this can be of interest for scientists. As for the temperature influence, the matrix-dominated properties undergo a significant change at the temperatures above T_g . Therefore, the latter does not seem to vary with absorbed moisture. The current review eventually serves to establish the test matrix for experimental characterisation of hygro-thermo-mechanical behaviour.

I.1.6. Conclusions

The aim of this research work is to analyse the environmental effects on the mechanical response of bolted joints with clamped composite parts. The storage and use of the proposed composite materials may be affected by climatic conditions, especially in the automotive domain where the working temperature and relative humidity vary respectively from -40°C to +80°C and from RH 0% to RH 85%. Hence, the characterisation of bolted joint behaviour also necessitates a complete database of composite material properties, mainly due to their important sensitivity to mentioned environmental conditions. Therefore, the proposed literature review reveals a great number of already studied aspects and those to consider in this work. As described above, these are the physical, chemical and mechanical properties under extreme thermal and humid conditions. They are particularly essential by reason of the application fields of the materials and not full availability in the open access. In addition, the creation of a material database also requires the results under intermediate environmental conditions. Thus, the literature review demonstrates how and what changes may cause the constantly fluctuating humidity. The hygroscopic PA6 GF and PA6 undergo the most important changes. Little quantity of moisture is absorbed by PPS CF; thus, its mechanical behaviour is generally unaffected. As for the exposure to temperatures, it results in a significant transformation of properties of both composite materials. When these conditions, moisture and temperature, act together, their coupled influence leads to a decrease of tensile, compressive, flexural and shear strengths, higher creep compliance, increased strains, lower moduli. These changes are especially visible in the vicinity of their glass transition temperatures. In addition to reversible physical damage, prolonged exposure to such an environment may also generate irreversible chemical damage. Nevertheless, the main focus is further attributed to physical and mechanical impact due to relatively short exposure to a hostile environment.

I.2. CHARACTERISATION OF BOLTED COMPOSITE JOINTS

The following characterisation of widely used bolted joints clarifies the benefits for industrial applications in comparison to the other composite-metal joining methods. Occurring solicitations and associated failure modes are analysed in relation to possible hygro-thermal conditions. The importance of preload application, its impact on the mechanical behaviour of joints over time and its dependence on the joining materials are particularly highlighted in the section. One can also unveil in the section why the application of bolted assemblies, joining composite materials that are sensitive to temperature and moisture, requires a thorough understanding and computation of their mechanical behaviour and capability to sustain external loading.

I.2.1. Review of composite-metal joining methods

A necessity to employ different materials in a structure, containing numerous parts, leads to the use of various connection types that are to be safe. They can be either demountable or permanent; hence, one is preferred to the other, or they are combined according to an application field, a structural material, external loadings, etc. The fundamental role of joints is to transfer loading within a structure. What is more, they are potentially the weakest elements; thus, it is important to minimise their quantity. It would, eventually, minimise the weight and cost, that is one of the principal criteria not only in aviation but also in automobile industries due to CO_2 restrictions. To make it efficient, appropriate joint types are to be selected in accordance with further applications and employed materials. Some specific assemblies had been developed for composite materials in particular; however, a general brief review is present in this section.

Adhesively bonded joints

Development and application of new matrices significantly contributed to the growth of bonded assemblies. The particularity of this conventional joint type is that the applied load is transferred by shear on the adherents – joined elements [128]. Hence, the failure can potentially occur due to: shear, tension or compression of adherents; shear or peel in the adhesive layer; adhesive failure at the composite/adhesive interface; shear or peel in the composite near the interface. For a good design, both the elements to join (a type of used material, geometry, size, potential failure modes, compatibility with polymer matrix) and in-service conditions (external loading, working environment) must be accounted for [19,129,130].

Advantages	Disadvantages
Small stress concentration	Limits to thickness that can be joined
Stiff connection	Difficult inspection
Good fatigue properties	Possible environmental degradation
Sealed against corrosion	Sensitive to peel and through-thickness stresses
Smooth surface contour	Residual stress when joining to metals
Relatively lightweight	Cannot be disassembled
Damage tolerant	Requires high degree of quality control
No fretting problems	May require costly tooling and facilities
	Risk of corrosion
	Time-consuming

Table I-4: Advantages and disadvantages of adhesively bolded joints [19,128,131]

As any other joint type, adhesively bonded joints possess both advantages and drawbacks, introduced in Table I-4, that define the efficiency for every application type (Figure I-73). Environmental conditions are,

therefore, one of key parameters due to highly probably hygro-thermal ageing of the adhesive layer as cited in section I.1.2.

Along with disadvantages, the benefits of adhesive bonding led to extensive use in aircraft and automobiles [132]. What is more, the combination of adhesive and mechanical joining, for instance, fastener reinforced adhesive joints as cited by [128], broadens the scope of application. Among these techniques, we can cite [128,133]:

- fit form: joining as a part of the curing cycle;
- pre-cure fastener installation;
- comeld technology.

More details regarding the adhesive assemblies are cited by Hamonou [1].



Figure I-73: Classification and application of adhesively bonded joints used in airframe manufacture [19]

Welded joints

Weld-bonding can be seen as an alternative technology of combined conventional adhesive and welding methods and, thus, possessing advantages of both. This method is attractive for high-performance automotive manufacturing, for instance, where the composite underbody is joined to steel body-in-white as shown by Shah et al. [132] (Figure I-74 – Figure I-75). In order to cure adhesive, the steel and the composite are held together with resistance spot welding, which also serves as an alternative load transfer, as explains Joesbury [128]. According to Darwish et al. [134], there are two approaches to assemble: the flow-in and the weld-through techniques. In the first case, the parts are firstly welded, then the adhesive is applied followed by curing; in the second the adhesive is applied first with the consequent assembling, then parts are spot-welded and heated.



Figure I-74: Weld-bond joint used to join composite to steel [132]

Figure I-75: Weld-bonded joint concept [132]

Santos et al. **[135]** investigated numerically and experimentally the mechanical behaviour of welded joints with epoxy and methacrylate matrices. The authors demonstrate that, in general, weld-bonded joints with epoxy show better performance than simple spot-welded; however, it is not the case with methacrylate. Hence, in this

particular case the thermoset matrix is preferable to thermoplastic. Nevertheless, peel strength remain similar due to the dependence on weld nugget and not the adhesive. It is also confirmed by Shah et al. **[132]** resulting from the quasi-static and fatigue tests at different temperatures. According to their results, the weld-bonded joints considerably outperform adhesive joints. The requirements for a weld-bonded joint possessing good performance are cited in **[132,134,135]**.

Z-pinning and CMT pins

Z-pinning and cold-metal-transfer CMT pins represent relatively recent methods that are under examination for many years by now. However, they do not take over the conventional joint types in industrial applications.

Z-pinning allows reinforcing prepreg laminates through the thickness, as demonstrated in Figure I-76 and Figure I-77 **[136–138]**. Z-Pins can be manufactured of titanium alloy, steel or fibrous carbon composite **[139]**. A principal concern is that the pins may damage a composite structure on a micro-scale due to their higher rigidity as compared to prepreg. Advantages and drawbacks of z-pinning are summarised in Table I-5.



Figure I-76: Schematic architecture of the foundation arrangement for a z-pin in: a) UD laminate; b) Quasi-Isotropic laminate [137]



Figure I-77: Cross-section view of a z-pin in UD laminate. The pin is inclined at an angle from the orthogonal direction [138]

Advantages	Disadvantages
Improved delamination toughness	Reduction of in-plane mechanical properties
Higher impact resistance	Complicated quality control
Damage tolerance	Difficulties for accurate insertion
Increased through-thickness stiffness	Fibre damage
Better joint strength	Swelling of the laminate
Improved bearing strength	Degraded durability
Common prepreg manufacturing process	Fibre distortion and crimping near z-pins
One-side access to the mould	Formation of matrix-rich zones
Better fatigue shear strength	Prone to crack initiation

Table I-5: Advantages and disadvantages of z-pinning. Inspired by [139]



Figure I-78: A method of joining by means of embedded longitudinal fastening microelements [140]

CMT pins propose an alternative method for metal-composite joining as cited in [1,128]. They are represented as fasteners embedded in a metallic sheet. The pins can equally reinforce a composite if accurately designed and inserted. What is more, such a method has strong future prospects in contrast to conventional

joining techniques by reason of maintaining the integrity of a composite. CMT pins enable in-plane (Figure I-78) and through-thickness connections (Figure I-79).

According to Karpov [140,141], the embedded metallic fasteners can be of different geometry or positioning (Figure I-79). This technique can also be combined with adhesive bonding if pins are covered with an adhesive layer.

Another novel and interesting bonding design with the use of pins, named half-stitch joint, was developed, studied and implemented in [140,142] (Figure I-80). This complex metal-composite heterogeneous structure provides an efficient transition from a composite to metal in delicate areas in particular. What is important to note is that the half-stitch joint allows controlling the fibre/yarn positioning significantly reducing their possible damage. According to [140], the main difficulties consist in an appropriate material winding in order to limit residual stresses and to control tension force; in maintaining equal centre-to-centre distance to avoid yarn slackening; and in keeping the designed fibre volume fraction in the tip zone.



Figure I-79: Design and engineering solution for joining by means of embedded transverse fastening microelements [140]



Figure I-80: Modified half-stitch jointing [140]

Riveted joints

These mechanical joints were initially developed for metal-to-metal connections standing out for their rapidity and mechanical properties. They are still widely applied in structures requiring light weight (for instance, in some airplane parts). Rivets are also used in composite-metal connections. An approximate process configuration is illustrated in Figure I-81.



Figure I-81: Schematic view of the FricRiveting process in metallic-insert joints, (A) positioning of the joining partners, (B) feeding of the rivet into the polymer (friction), (C) rivet forging, and (D) joint consolidation [143]

Many authors studied the application of riveted joints for composite materials. Li et al. [144] analysed the static and dynamic behaviour of single-lap and double-lap riveted carbon-fibre composite. They discovered that

the failure modes vary with loading rates and specimen symmetry: bending failure, longitudinal penetration and rivet rotation were identified along with coupled bearing/pull-out and bearing/cleavage. Authors state that the joint failure is progressive when loading is of constant velocity; however, the failure is immediate when used in a pre-tensioned structure. Effect of heat treatment, similar to the manufacturing process of vehicles, on composite-metal joined test specimens was studied by Kang et al. [145]. They highlight that the top ply of composite panel around the rivet, as shown in Figure I-82, can lead to bearing failure. Overall, good static and fatigue resistance was achieved by [143,146,147].



Figure I-82: Representative cross-section of untested self-pierce riveted carbon fibre reinforced plastic (CFRP) - aluminium test specimen; (a) macrograph showing the CFRP and aluminium sheet interlocked by a steel rivet; (b) and (c) magnified view of the rivet and aluminium surface interface under SEM showing the presence of a thin layer of CFRP [145]

Advantages and disadvantages of riveted composite-metal joints are outlined in Table I-6 according to the literature review.

Advantages	Disadvantages
No heating involved	Localised fibre damage
No pre-drilling holes	Deformation of the bottom sheet
No surface preparation	Stress concentration around the rivet
Hermetically sealed	Change in mechanical properties around the rivet
Short joining cycles	Cannot be disassembled
Low-cost machinery	Only spot-like joints
High mechanical performance	Closing force is not readily controlled
Superior fatigue life	Restriction on the minimum allowable thickness
Full shearing of upper sheet	

Table I-6: Advantages and disadvantages of riveted joints, inspired by [143,145,148]

I.2.2. Bolted composite joints

The necessity to assemble composite and metallic materials is caused by the use of large structures or its complex geometry, the demands of optimisation of composite material use, the cost reduction, etc. [149]. Regardless of the constant developing of brand-new and promising joining types dedicated to composite materials, bolted joints remain among the most commonly used. Bolts belong to mechanical fasteners used for structure assembling in a vast and diverse range of domains. They offer the greatest strength for mechanically fastened composite structures. The performance of bolted joints is related to a great number of parameters that are briefly discussed in the present section. The introduction to the solicitation and failure modes, to the relevance of preload and its relation to composite materials can be found in this section as a part of the literature review and in Chapter IV.

I.2.2.1. Introduction to bolted connections

The fastening parts of a bolted joint can be of different diameters, pitch of thread, property class, finishing etc. and they are standardised. Types of mechanical fasteners are schematically illustrated in Figure I-83. However, there exist many categories of bolts (Figure I-84) that propose different competitive solutions as explored by Heistermann [150].



Figure I-83: Types of mechanical fasteners. From left to right: bolt; screw; stud; screw in the threaded bush [151]



Figure I-84: From left to right: Tension Controlled Bolt; Huck BobTail lockbolt; Standard bolt in combination with NordLock washers; Friedberg HV Randel [150]

One can see in Figure I-85 a cross-section of a typical bolted joint. Usually, it consists of a bolt, two washers, two clamped materials and a nut. Washers are not mandatory, but essential for composite joints since they help to uniformly distribute the applied loading. What is more, a clearance between joint materials and a bolt is generally present [152,153].



Figure I-85: Joint nomenclature [152]

Detailed mechanical behaviour of bolts under tension, compression, shear and their combinations at rest and during installation is described by Kulak et al **[154]**. As for the composite joining, its peel failure is not an issue; however, low bearing and transverse strength may negatively contribute to the joint behaviour. Similarly, material disarrangement and damage due to a clearance hole, creep damage over time represent additional problematic related to composite materials in bolted joints **[19]**. However, the possibilities to maintain good performance over time and to filter external forces are not neglected; thus, the bolted connections are widely used to fasten the composite materials. Coupled, bolted and adhesively bonded, joint leads to even higher performance than a

single mechanical joint. Main advantages and disadvantages of bolted joints are cited in Table I-7. Despite being a source of stress concentration, bolts remain reliable, improve structural efficiency and maintainability **[155]**. Another relevant benefit, differing bolted from the other mechanical joints, lies in the capability to apply a preloading force that would delay the separation of assembled parts (section I.2.2.3).

Advantages	Disadvantages
Can be disassembled	Considerable stress concentration
No thickness limitations	Hole formation can damage composite
Simple joint configuration	Poor bearing properties of composites
Simple manufacturing process	Prone to fatigue cracking in metallic component
Simple inspection procedure	Environmentally sensitive due to composites
Provides through-thickness reinforcement	Prone to fretting in metal
No major residual stress problem	May require shimming

Table I-7: Advantages and disadvantages of bolted composite joints [19]

Bolts can be found in a lot of surrounding structures, like bridges, mechanisms, items. For a particular application, special bolts can be designed as for a composite wheel for instance or for aircraft applications.



Figure I-86: One-piece carbon composite wheel1



Figure I-88: Section of air intake of A350. An acoustic panel of sandwich structure ³



Figure I-87: Rib of the rudder of A400M. Carbon-fibre reinforced thermoplastic composite²



Figure I-89: Section of air intake of A350. An acoustic panel of sandwich structure ⁴

¹ https://www.designnews.com/materials-assembly/one-piece-carbon-composite-wheel-drives-cars/69959761132388

² From Technocampus Nantes

³ From Technocampus Nantes

⁴ From Technocampus Nantes

I.2.2.2. Solicitation and failure modes

An accurate design of bolted joints is determined by many parameters that one has to take into consideration. The choice consists not only in a type of bolt and its quantity, positioning, a material it is made of, but also the in-service environment and solicitation modes, non-critical failure types, preload to apply (section I.2.2.3). Typical solicitations are demonstrated in Figure I-90. When a joint is subjected to external acting forces, the combination of the loading types can occur. For instance, in an eccentrically loaded bolted joint a torsional moment and a shear force appear [154].



Figure I-90: Loading types [156]

The tensile loading on bolted joint mainly leads to tension failure of a neat section, bearing, shear failure and some mixed-mode failures: cleavage/tension, tension/shear, bolt failure due to bearing, bolt-head pulling through the laminate **[19,148,155,157]** (Figure I-91).

Numerical and analytical failure strength and failure mode predictions are cited by Chang et al. [158], by Ramkumar et al. [155], by Matthews et al. and Chang et al. [157] for different laminate layups. The proposed extra design criteria related to the joint configuration is its location within a structure, as cited in [155]. Computational approaches for bolt mechanical behaviour under different loadings are proposed in [151].



Figure I-91: Schematic illustration of the main failure modes in mechanical joints of composites by Hart-Smith [157]

Bolted TP and TS composite joints with single and double lap were intensively investigated by Vieille et al. **[43]**. The authors also demonstrated the effect of environmental conditions – dry at room temperature and humid at +120°C – on the joint mechanical response. Apparently, in the case of the double lap, the latter condition changes the failure mode of PEEK CF laminates from shear-out to bearing, whereas in PPS CF and Epoxy CF the failure is shifted from cleavage to bearing. Such behaviour can be explained by a ductile nature of matrices at high temperatures. Due to the asymmetry of single lap joints, a bolt-hole clearance and hostile environment, out-of-plane plastic deformation is dominant (Figure I-92). Analogous results are attained in **[110,118]**. More details regarding the mechanics of bolted joints can be found in **[1]**.



Figure I-92: Failure surfaces at room temperature (on the left) and +120°C (on the right) of a single lap joint [118]

Subsequently, mechanical properties and failure mechanisms of bolted joints have a strong tendency to vary with clamped composite materials. Type of composite matrix and its state, ductile or rigid, plays an especially important role for failure modes. The other laminate parameters, as a reinforcement type and stacking sequence, also define the sort of failure for in-plane loading. The nature of composites is such that the working hot or humid conditions can be a key-point to appropriate design of bolted composite joints for the reason that the high compliance of assembled materials and a tendency to relaxation over time can significantly affect also the out-of-plane joint behaviour (sections I.1.4 - I.1.5).

I.2.2.3. Notion of preload

Importance of preload

One of the decisive parameters determining the joint ability to withstand applied external load is the appropriate pretension force. It is thus important for bolts to maintain it over time under working conditions. According to **[1,150,152,159,160]**, many factors shall be taken into consideration: the type of clamping force, fatigue concern, maximum applied torque, yield strength of bolts, materials of bolt and clamped parts, load introduction factor, load position, filtering coefficient etc. Most widely used tightening methods are cited in **[1,156,160]**.

In Figure I-93 on can see the schematic behaviour of a bolted joint without and with pretension. In the first case, a joint would fail in a matter of seconds once the external loading is applied since $F_b = F_{ext}$ (black dashed line). When some pretension is initially applied to a bolt, F_0^b , the failure of joint occurs at the moment of intersection of the blue and the black dashed slopes with the consequent $F_b = F_{ext}$. The illustrated graph represents both the main interest and main challenge when employing bolted composite joints.



Figure I-93: Schematic representation illustrating the differences in behaviour of a rivet and a bolt in the case of centred axial loading [1]¹

¹ Fixation stands for fastening; Boulon stands for bolt; Ligne symbolisant le décollement stands for Line representing the separation

Application of preload

The preload application results in a compression of assembled members and an elongation of a bolt (Figure I-95). The washers, if used, distribute the pressure on the part surfaces equivalent to their diameters. Their use is of particular interest with composite materials due to the low bearing strength of the latter. The deformation cone occurs in the parts; it is, therefore, integrated in the computation methods and the standard **[159,160]**.

The relation between the tensile load in a bolt and external forces is established via the filtering coefficient λ that is dependent on the compliance of a bolt δ_b and assembled parts δ_p [159,160]:

$$F_b = F_0^b + \lambda F_{ext} \tag{2}$$

$$\lambda = \beta \frac{\delta_p}{\delta_b + \delta_p} \tag{3}$$

$$\delta_b^i = \frac{l_b^i}{E_b A_b^i} \tag{4}$$

$$\delta_p = \int_{z=0}^{z=L_p} \frac{dz}{E(z)A(z)} \tag{5}$$

From the equations above we can see that the type, rigidity and geometry of assembled materials play a crucial role in determining the pretension force and joint ability to filter the external forces.

It is interesting to note that when a preload is applied with torque, a bolt undergoes both tensile and torsion stresses (Figure I-94). This obviously affects the equivalent stress in the bolt. Therefore, both stresses are to be taken into consideration for preload computation in order to remain below the bolt yield strength **[156]**.



Figure I-94: Torsion stresses in the bolt resulting from torquing **[156]**



Related issues

One of the principal challenged of bolted connections is the self-loosening over time, which leads to the necessity of regular maintenance. It is directly related to the decrease of applied preload, part creep, bolt plasticisation in the critical parts. For instance, the loss can reach 4-7.5% in the case of metallic assembly with applied coatings **[150,161]** (Figure I-96). Higher preload loss is expected in joints with the use of composite materials. It is related to the weak resistance to creep over time, smaller stiffness as compared to bolt, which can be aggravated by the impact of environmental conditions (sections I.1.4-I.1.5).

The decrease of applied preload occurs in three distinct phases and is accurately described by Heistermann [150]. Right after the tightening and up to 10 s, one can observe the initial preload loss, which is mainly dependent on the clamping method. According to the author, the immediate relaxation is also higher if the tightening is faster or tightening force is higher. In the case of composite members, the adjustment of rough

¹ Etat initial/serré stands for Initial/tightened state; Pièce stands for part; Frottement boulon-rondelle pièce 1 (2) stands for Friction bolt-washer part 1 (2); Compression des pièces 1 et 2 stands for Compression of parts 1 and 2; Allongement du boulon stands for bolt elongation; Frottement écrou-corps boulon stands for friction nut-bolt shank;

surfaces may occur. The short-term relaxation occurs in the first twelve hours. The preload, applied to the bolt, induces the compression in the assembled parts through the nut, which tend to deform plastically due to the outof-plane creep of composites. Consequently, the initial bolt elongation starts to reduce. The short-term relaxation is followed by the long-term one that can actually be described as its asymptotic extension. What is interesting to note is that the tightening of a group of bolts causes their mutual interaction: while tightening second bolt, the first undergoes the simultaneous loss of preload.



Heistermann **[150]** predicted the loss of pretension for twenty years as a function of a number of coating surfaces (Figure I-97). It is clear that the thickness of the deformable part of specimens is of absolute importance for preload retention. We can presume that this impact can be higher for bolted composite joints via the filtering coefficient and part compliance due to an important deformable thickness.



Figure I-97: Average bolt forces of short Huck bolts depending on the surface finishing extrapolated for 20 years in comparison with EN 1993 **[150]**

I.2.2.4. Dimensioning of bolted joints in CETIM-Cobra

As mentioned in the previous sections, the design of bolted joints is directly related to the properties of assembled parts and bolt, applied forces, etc. A European standard VDI 2230 with extensions NF E 25030-1 and NF E 25030-2 **[159]** specifies the integral computation of such connection, providing a complete mechanical behaviour and a final product design. The methodology, firstly introduced in 1977 and then revised in 1986, 2001 and 2015, introduces numerous parameters, used for the appropriate design. The computation of bolted joints is based on the mechanical characteristics of its elementary constituents. However, computation, design and optimisation of bolted joints require a lot of working hours. In order to facilitate this and save time, the CETIM developed and commercialised the software CETIM-Cobra, which is based on the standard. It is nowadays adopted by diverse enterprises, specialising in aeronautical, automobile, railways, oil and gas domains.

¹ Temps stands for time ; Assemblage stands for assembly ; Charge de serrage stands for tightening load

Based on the parameters of elementary constituents, the software verifies the clamping forces, static, dynamic and thermal operating conditions. For complex cases, the algorithms include more than 60 different computations. The software includes 8 databases of standards: ISO, EN, AFNOR, DIN, BS etc. [162]. Nevertheless, the software, as well as the standard [159], is yet limited to the use of metallic materials in terms of a homogeneous structure and elastic properties. Resulting from the impact of humid conditions on the composites, the absence of moist operating conditions is an important drawback. Increasing use of bolted composite joints requires the extension and adaptation of software.

I.2.3. Conclusions

Many methods for joining composite materials to metals have been developed and successfully employed so far. These are adhesively bonded joints, welded joints, z-pinning, CMT pins, riveted and bolted joints – this list is not exhaustive. Nevertheless, the bolted joints remain among the most widely used for composite-metal connections due to their particular features. One of the most important is the possibility to delay the joint failure by applying preload that generates elongation of bolt and compression of clamped parts. The applied prestress defines the joint capacity to withstand acting external forces; therefore, the prestress should remain below the bolt yield strength. In addition, the mechanical behaviour of bolted joints is also related to a number of parameters of elementary constituents, working conditions, loading type and its position. Among the elementary constituents, one can cite a bolt, a nut, washers and clamped parts. The latter, if made of composite material, is of special significance due to generally lower out-of-plane stiffness, creep resistance and bearing strength in comparison to metals. Its high compliance may cause more rapid spontaneous loosening and overall higher preload loss. The working conditions, represented by humid and thermal impact, lead to aggravated degradation of the material itself as demonstrated in the previous sections; thus, more rapid preload loss is possible to occur. As for the loading conditions, both dynamic and quasi-static influence may result in different failure modes of bolted composite joints: tension and bearing failure, shear-out and cleavage, bolt failure, etc. Hence, the literature review outlines the problematic of the present research, namely, the evolution of applied preload to bolted connections with composite clamped members as a function of environmental conditions.

I.3. RESEARCH INTERESTS

An intensive application of composite materials with superior mechanical and physical properties leads to a necessity to expand the software CETIM-Cobra, based on the widely used in Europe standard VDI 2230 with extensions NF E 25030-1 and NF E 25030-2 and adopted by French enterprises. Constant investigation of the sensitivity of certain composite materials to particular environmental conditions and a clear understanding of their changing characteristics may eventually result in a requirement to involve humid and environmental impacts for better joint design. The behaviour of bolted joints, integrating composite materials, has been studied by R. Hamonou [1] that constitutes the first phase for the clear and rigorous methodology for application of woven composite materials as clamped parts. The proposed numerical approach for the definition of compliance of the heterogeneous structure of assembled materials is based on the energy criterion used for finite element simulations. The author also advanced the analytical model for the determination of "filtering" coefficient that defines the behaviour of preloaded bolted joint. In the present work, the suggested numerical approach is adopted with the use of environmental conditions. As already mentioned, the out-of-plane properties of joint members, characterising their compliance, are of first concern in the case of prestressed bolted connections. Hence, a rigorous evaluation of the study materials is of absolute importance prior to the analysis of joints. Weak resistance to creep, low elastic properties, and hygro-thermal influence on the parameters of clamped parts may also cause faster and more important loss of applied preload and lower resistance to fatigue loadings. In addition, the database of loss of preload in bolted composite joints is rather weak in comparison with metal joints. Hence, the lack of such information at the varying environment is the major motivation to realise the research work.

As demonstrated in the literature review, a significant effect of surrounding temperature and humidity, acting separately or simultaneously, on proposed composite materials results in the degradation of physical and mechanical properties over time. It is particularly important for parts employed in harsh in-service conditions. Despite a large number of published papers on the environmental impact on composite materials, the lack or absence of a consistent methodology for the experimental characterisation under hygro-thermal impact in open access represents a drawback for industrial and scientific fields. To master it, an approach for the elaboration of a conditioning protocol and its application for a rigorous in-plane mechanical characterisation of the materials are proposed in Chapter II. This campaign provides a database of elastic mechanical properties of PA6, PA6 GF and PPS CF as a function of the moist and thermal atmosphere. These results equally serve for identification and validation of a whole range of numerical parameters at the same environment with the use of reconstructed RVE of a woven structure as presented in Chapter III. This campaign is necessary for the reason that the out-of-plane properties of composites, which define the mechanical behaviour of bolted joints over time, are not easily accessible via experimental approaches. Besides, obtained properties finalise the material database. Hence, a double-scale homogenisation approach is employed for their rigorous characterisation via finite element method. Obtained out-of-plane elastic moduli serve for the estimation of compliance of clamped parts, made of a composite, with analytical and numerical methods in Chapter IV. The accomplished experimental and numerical evaluation of material properties allows the computation of compliance for different environmental conditions. The evolution of preload, affected by the member response and structure, is likewise studied. These results are expected to be integrated into the software CETIM-Cobra in order to manage the sensitivity of the composite materials to given environmental conditions. Finally, the general conclusions of the research work are presented in the last section along with perspectives.

CHAPTER II

EXPERIMENTAL CHARACTERISATION

The principal objective of the present research work is to analyse the mechanical behaviour of prestressed bolted joints with composite members. The interest lies in characterising the change of compliance of clamped parts and the loss of preload over time under certain environmental conditions. In order to perform it, the mechanical properties of employed composite materials should be known at the same environmental parameters. Therefore, the methodology for the experimental characterisation is proposed and implemented in the present Chapter. Initially, an absorption/desorption protocol is developed for the composite PA6 GF with the consequent validation for the neat matrix PA6 and the composite PPS CF. The conditioning protocol is applied to all the experimentally characterised specimens. During the experimental campaign, we particularly seek the elastic properties of the materials that provide an important database for varying in-service environment. Besides, they are afterwards used for numerical simulations of a Representative Volume Element of the composite material PA6 GF (Chapter III) and evaluation of compliance of clamped parts (Chapter IV). Every section is dedicated to a specific task and finishes with brief conclusions.

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II.1. DESCRIPTION OF MATERIALS AND SPECIMENS

This section presents a review of three provided thermoplastic materials: polyamide 6, glass-fibre reinforced polyamide 6 and carbon-fibre reinforced polyphenylene sulphide (the neat matrix PPS was unavailable at the moment of this research work). Due to the various experimental campaigns, twelve specimen types are used for characterisation of given materials. Specimen geometries vary for each testing method: a dog-bone specimen type of the matrix is used for the validation of conditioning protocol, determination of out-of-plane hygroscopic deformation and mechanical experimental campaign; the rectangular matrix specimens are principally used for the evaluation of hygroscopic deformation. As for the composite materials, the specimens are of three fibre orientations 45°, 0° and 90° of rectangular and dumbbell-shape geometries. They are employed for the conditioning protocol development, analysis of in-plane thermal and hygroscopic expansion and mechanical experimental in-plane tensile testing.

II.1.1. Brief review of materials

PA6

The thermoplastic material polyamide 6 is manufactured with injection method and provided in the form of standard specimens of different geometries with 4 mm thickness. Although no extra information is available, one can assume the neat resin properties correspond to the matrix of the composite material PA6 GF presented below.

PA6 GF

Thermoplastic composite PA6 GF stands for the polyamide 6 reinforced with continuous roving glass fibres of a trademark Tepex® Dynalite, supplied by Bond-Laminates. Laminate is delivered in the form of plates of three total thicknesses: 2 mm, 4 mm and 6 mm. The thickness of each layer is 0.5 mm, thus the number of layers is 4, 8 and 12 respectively. According to the supplier, the fibre volume content is 47%. The composite material is stated to be quasi-balanced for the reason that the reinforcement is a fabric of a twill weave 2×2 and it has an equal weight rate in both longitudinal and transverse orientations with 1200 tex.

Regarding the manufacturing, the laminate is processed by a press forming method in 2 steps. The first phase consists in brief heating in the range of $+240^{\circ}\text{C} - +260^{\circ}\text{C}$ and it is followed by the forming, where the closing speed of the press is superior to 50 mm/s at the beginning and is reduced to 5 mm/s for the last 10 min of forming. Then the cooling takes place at a temperature lower or equal to $+110^{\circ}\text{C}$ under a consolidation pressure during about 30 s according to the material datasheet [163].

PPS CF

The composite material PPS CF represents the continuous carbon fibres providing the reinforcing properties to the polyphenylene sulphide matrix. The material is likewise registered under the trademark Tepex® Dynalite of Bond-Laminates. The thickness of laminates is 1.5 mm with 0.25 mm per layer. The fibre volume content is expected to be 45%. Similarly, the type of reinforcement is a twill carbon fabric of 2×2 with areal weight 200 g/m² and 3000 yarns in longitudinal and transverse directions. Such reinforcing provides the quasi-balanced properties as shown in the material datasheet [164].

As for the processing, the laminate is made with a press-forming method. The material is heated up to $+310^{\circ}\text{C} - +330^{\circ}\text{C}$ on two sides. Then the press forming takes place at similar conditions as PA6 GF; however, no precise information is given. The last step is cooling under a consolidation temperature as mentioned in [164].

II.1.2. Specimen preparation

Specimens, used in the present work, are of several geometries due to different objectives of the experimental campaigns. As for the composite materials, they were cut out with a water jet according to demanded dimensions and fibre orientation. All the specimens were then referenced and stored in ambient environmental conditions. After the desiccation/absorption procedures (section II.2), the specimens were placed into sealing bags according to the testing batches (section II.4).

II.1.2.1. Neat matrix

Three specimen types of the neat matrix are used in this research work: a dog-bone (Figure II-1) and two rectangular (Figure II-2 – Figure II-3). The dog-bone specimens are used for the characterisation of moisture content (section II.2) as well as for the mechanical characterisation in the experimental campaign (section II.4). The dog-bone and rectangular specimen types are employed for the analysis of hygroscopic swelling in order to have the consistency of results. The total number of tested dog-bone specimens is 105 and 12 for rectangular.



II.1.2.2. Composite samples

All the specimens, except circular, are cut out from the laminates at 45°, 0° and 90°, produced within one manufacturing process for each composite. Hence, no differences between specimens of one material, caused by processing, are expected. The composite specimen geometries are illustrated in Figure II-5 – Figure II-9.

45°-fibre orientation

In terms of application, the specimens of 45°-fibre orientation are used for the elaboration of a desiccation/humid ageing protocol for PA6 GF (section II.2) and its adjustment in the case of PPS CF, the determination of in-plane shear properties in the tensile testing campaign (section II.4), and the thermal expansion tests (section II.3). Regarding the protocol, tensile testing is one of the necessary experiments to select the conditioning temperature. This particular fibre orientation of specimens may reveal changes in the matrix due to the matrix-dominated properties.

ISO 527-5 gives the recommendation of the specimen geometry for UD composite materials. In the case of the woven composites, no recommendation is provided. Nevertheless, it is strongly suggested to take the size of a **R**epresentative **V**olume **E**lement (RVE) into consideration, thus at least 1.5 of an RVE should be in the effective width. An RVE here is the smallest volume of a composite material that can provide the properties of an entire material when tested. The rotated RVE is proposed for 45°-fibre oriented specimen, which is basically

smaller than that of 0° and 90°-fibre oriented specimens. Therefore, the chosen specimen width (Figure II-5) is nearly 2.4 of the RVE with dimensions $10.5 \times 10.5 \text{ mm}^2$ for PA6 GF and nearly 4.2 of the RVE with dimensions $6 \times 6 \text{ mm}^2$ for PPS CF if one considers the RVE, rotated at 45° (Figure II-4).



Figure II-4: Red rectangle depicts the RVE for 45°-fibre oriented specimens of a) PA6 GF and b) PPS CF

All the specimens, dedicated to mechanical testing, are initially subjected to the desiccation or the desiccation/absorption. Adhesive, used to attach the end-tabs, is probable to be sensitive when subjected to a humid environment. It may lead to the sliding between a specimen and the end-tabs during traction. In order to prevent the side effects, it is decided to renounce the end-tabs despite the suggestion of ISO 527-5.



Figure II-5: Rectangular composite specimen with 45°-fibre orientation. Type 4

0° (warp) and 90° (weft) fibre orientations

These specimens are used in a tensile testing campaign on PA6 GF and PPS CF; it is, therefore, necessary to ensure the breakage in the effective length. However, the specimens of 0° and 90°-fibre orientations and the standard geometry (Figure II-8) are known for the rupture near the grips due to the occurring stress concentration, which depends on numerous parameters as stated by **[165–167]**. Another challenge lies in the absence of the material ability to the reduction in the area during tensile testing. It is caused by a relatively small Poisson's ratio as also pointed out by **[165]**. In order to localise the rupture in the centre of specimens, the curved configuration is thus selected. Owing to the woven composite structure, the pull-out failure might occur during testing as reported by **[165]**, but no relationship is established between the transverse and longitudinal failures. Hence, the specimens of 0° and 90°-fibre orientations are of a dumbbell-shaped geometry (Figure II-7) containing 1.5 of the RVE for PA6 GF and about 3 RVE for PPS CF. The size of the RVEs is estimated as 16×16 mm² for PA6 GF and 8×8 mm² for PPS CF (Figure II-6). The width of the narrowest area is restricted to the RVE of PA6 GF, whereas the width of 30 mm is defined by the grips, specially designed for testing in a thermal chamber. The absence of the end-tabs is justified by the desiccation/absorption processes as in the case of 45°-fibre oriented specimens.

A few specimens of PA6 GF, illustrated in Figure II-7, are used for the development of the conditioning protocol (section II.2). The rectangular geometry of PA6 GF and PPS CF specimens (Figure II-8) is used for the analysis of thermal expansion.







Figure II-7: Dumbbell-shaped composite specimen with 0° and 90°-fibre orientation. Type 5



Circular samples

The use of circular specimens is mainly related to the experimental and numerical study of bolted composite joints in Chapter IV. Therefore, they also serve for the protocol validation and evaluation of hygroscopic out-of-plane deformation for different thicknesses of PA6 GF. Laminates of 2 mm and 4 mm thickness were delivered simultaneously and stored at the same conditions. The sheet of 2 mm comes from the same batch as the one for 45°, 0° and 90°-fibre oriented specimens as described above. As for the laminate of 6 mm, it was manufactured 4 months before the onset of experiments. The content of RVE in the specimens in relation to their surfaces is 3.976 for the diameter 36 mm and 0.994 for the diameter 18 mm.



Figure II-9: Circular composite samples of two diameters (a-c, d-f) and three thicknesses (a, d), (b, e), (c, f). Type 7

II.1.3. Conclusions

Two thermoplastic composite materials and one neat matrix PA6 are selected for the research work due to the unavailability of neat PPS. The mechanical properties and the moisture content of the specimens are unknown due to the prolonged storage of laminates under unregulated environmental conditions with the exception of laminates of 6 mm thickness. The latter are manufactured 2 months prior to the beginning of the experimental campaign. All the specimens are cut out with a water jet and marked according to the experiments types.

Concerning the selected geometries, the 45°-fibre oriented specimens serve for the establishment of the conditioning protocol, the in-plane mechanical testing campaign and the evaluation of a coefficient of thermal expansion. The dumbbell-shaped composite specimens are likewise used in the mechanical testing campaign. Several of them are, therefore, adopted for the development of conditioning protocol. The choice of circular

specimens is justified by their consequent applications for the tests on bolted joints. Besides, the out-of-plane hygroscopic deformation can be estimated by reason of important specimen thickness.

Rectangular and dog-bone matrix coupons are employed for validation of the proposed protocol. What is more, the dog-bone specimens serve for the mechanical characterisation of matrix properties.

II.2. ELABORATION OF CONDITIONING PROTOCOL

A clear and a consistent analysis of mechanical behaviour cannot be performed without using a suitable protocol for conditioning. It is especially the case for materials, extremely sensitive to environmental conditions. In the present section, we propose the step-by-step elaboration of an accelerated conditioning protocol for a composite material, sensitive to temperature and humidity. Importance of this procedure consists in the absence of such protocol with the following complete evaluation of conditioning temperatures, duration and impact on mechanical properties. Besides, the material manufacturer does not provide any information regarding conditioning. After the validation, this methodology is applied to prepare the specimens of PA6 GF for mechanical testing and evaluation the glass transition temperatures. Despite the low moisture uptake, the suggested conditioning method is adapted for the rigorous examination of hygroscopic properties of PPS CF that may have an impact on the elastoplastic material response, not studied in this work. The conditioning protocol is then applied to the PPS CF specimens for the mechanical testing campaign. The final humid ageing enables us to obtain mechanical properties of both the composites and the matrix under hygro-thermal conditions.

II.2.1. Concept of moist and moisture-free materials

Right after manufacturing, composite materials are supposed to be in a **D**ry-**A**s-**M**oulded (DAM) condition, which corresponds to the absence of water molecules within the polymer structure. Once materials are exposed to ambient conditions, they begin to absorb the moisture from the environment, which results in the change of physical, chemical and mechanical properties as cited in Chapter I and illustrated in Figure II-41. At some point, a complete saturation occurs at a given condition. For the research purposes, materials can be submerged in water at the selected temperature, placed into a climatic chamber with regulated relative humidity or simply kept outdoors to examine the impact of each condition. If reversible, material state can be returned back with the desiccation to the practically initial one. Nevertheless, a tiny quantity of water molecules remains trapped inside the polymer due to polymer chains/fibres/particles acting as a barrier. Consequently, the material in the dry state is close to the DAM, but is not absolutely equal to RH 0%. For the simplification purpose, we will use the dry material condition as a reference – or as a moisture-free material – as if it is constantly exposed to RH 0%.

II.2.2. Development of desiccation procedure for PA6 GF

Due to the prolonged storage in an uncontrolled environment, T_g is unknown due to the strong variation with the moisture content (section I.1.4.2). Supposing that the average RH during a year is over 65%, the current T_g should be less than +20°C, thus below the ambient temperature according to the study provided by [58]. The variation of T_g modifies the physical state of the laminate, and the degradation of mechanical properties can occur. Hence, it is important to avoid material damage during desorption and absorption. However, real working environmental conditions for automobile parts are such that the working temperature of a vehicle, supplemented by the RH, is quasi-exclusively above T_q , which varies nevertheless.

II.2.2.1. Temperature selection

According to the data-sheet of PA6 GF [163], its T_g is +60°C in the DAM condition. To accelerate desorption, the material should be subjected to the temperature higher than the ambient for a while [45,46]. This signifies that at the beginning of desiccation the composite, containing initially moisture corresponding to an environment around RH 65%, is in the viscous state, therefore, its T_g is small. Hence, the use of the desiccation temperature might be an issue for the mechanical material response. In order to evaluate the influence of drying temperatures on the kinetics of desorption and mechanical properties, three of them are selected: +50°C, +70°C and +90°C. Such a choice is governed and justified by T_g in the dry state. Thus, once the material reaches the minimum moisture content, the drying temperature +50°C remains below T_g , whereas +70°C and +90°C are

above. From a physical point of view, in the case of +50°C the composite is theoretically stiff at RH 0%, while at +70°C and +90°C it is in the viscous state. However, resulting from the vicinity of T_g , this statement may not be certain. Besides, the restriction of +90°C arises from the working temperature range of a thermal chamber.

For a greater desorption acceleration, all specimens are placed into a thermal chamber Binder VD-115 under vacuum (Figure II-10), otherwise, it might take months to obtain the moisture content close to RH 0% as observed in [4]. In addition, a short-term desorption process is necessary for manufacturers for rapid identification of material properties.

The moisture content is defined by a gravimetric method (ASTM D5229), cited by numerous authors [25,45,52,55,60,168], due to the absence of integrated balance in the chamber. It consists of weighing specimens on a regular basis. The precision of balance is 0.1 mg. Resulting from the comparison of the initial mass of a specimen, right before desorption, and at several time-steps, one can observe the change in mass due to the loss of moisture (Eq. 6). A major advantage of this method is the possibility to monitor several specimens at once. However, an inconvenience consists in the necessity of removing the vacuum and taking out the specimens for weighing. Even though it lasts barely 5 minutes, this manipulation might generate some errors in measurements. The gravimetric method still remains the most used one for the hygrometric study.

$$C_{glob} = \frac{M_t - M_0}{M_0} \cdot 100\%$$
 (6)

In the Eq. 6, M_0 represents the initial mass of a specimen, M_t the mass of the specimen at a time-step t (current mass) and C_{glob} the moisture content at a time-step t (current time). Once the moisture content reaches the plateau and stabilises, the specimen is assumed to be in the DAM state, or at RH 0%. Formulated differently, there is no moisture content in the material, or its amount is very little and negligible. Prolonged exposure may lead to continuous material degradation due to thermal-oxidative ageing (if the oxygen is present) [57]. However, within the framework of the current research, we assume that no chemical damage occurs due to relatively short desorption time, and C_{glob} corresponds to the value on the plateau.



Figure II-10: Thermal chamber Binder VD-115 with the possibility for vacuum connection

During 29 days of exposure at +50°C, the loss of moisture content is 0.797% without reaching the plateau (Figure II-11). Slightly higher moisture release is observed after desorption at +70°C during 14 days. And the maximum studied rate is at +90°C with attained stabilisation within 12 days. These results are in accordance with **[15,46]**, and the high temperature is assumed to be an activator for rapid desorption kinetics.

In order to validate the reproducibility of results, the mass monitoring is performed on six reference specimens of 45°-fibre orientation during 18 days (Figure II-12). On the 10th day the moisture content stabilises;

hence, 12 days of desorption appear to be sufficient. The maximum deviation from the average values of C_{glob} is $\pm 4\%$.



Figure II-11: Desorption of PA6 GF of 45°-fibre orientation at 50°C, 70°C and 90°C in vacuum



Figure II-12 Moisture content during desorption procedure of 45°-oriented specimens for tensile testing

II.2.2.2. Temperature validation through mechanical testing

Desiccated specimens subsequently undergo the controlled tension until failure on Instron 5584 testing machine at ambient temperature (+23°C). The strain field is assessed by applying a method of **D**igital Image **C**orrelation (DIC) (further explanation in section II.4). This step consists in determining the impact of the drying temperature on the mechanical behaviour and selecting the drying temperature for the desorption avoiding material damaging. Being matrix-dominated, fibre orientation of 45° with the geometry depicted in Figure II-5 is preferred here. Since the mechanical parameters of the woven composite material are obtained from the local stresses and strains (i.e. within the local frame, defined by the fibre directions), the transfer from the global frame

(i.e. defined by the tensile direction) to the local frame is achieved using the following equations (further explanation in section II.4):

$$\sigma_{xx \ eng} = \frac{F}{S_0} \tag{7}$$

$$\sigma_{12\,eng} = \frac{\sigma_{xx\,eng}}{2} \tag{8}$$

$$\gamma_{12 \, eng} = \varepsilon_{xx \, eng} - \varepsilon_{yy \, eng} \tag{9}$$

In the equations above, $\sigma_{xx eng}$ stands for the engineering tensile stress in global coordinates, $\sigma_{12 eng}$ represents the engineering shear stress in local coordinates, F is the applied tensile force, S_0 is the initial cross-sectional area of the specimen, $\gamma_{12 eng}$ is the engineering shear strain in local coordinates, $\varepsilon_{xx eng}$ and $\varepsilon_{yy eng}$ represent engineering axial and transverse strains in global coordinates. More detailed information about local and global frames is demonstrated in section II.4.3.1. The assumption, stating that the axes of orthotropy remain at 45° after tensile loading, is used here. This assumption can be false for large strains.

The impact of desorption temperatures is examined by carried tensile tests of 45°-oriented composite at ambient temperature (Figure II-13). This fibre orientation is selected for its matrix-dominated properties. Specimens are placed into sealing storage bags right after the previous desorption and then opened before the beginning of the current test in order to minimise the interaction with air.



Figure II-13 Tensile tests at the ambient temperature of PA6 GF specimens, dried at three different temperatures

Despite the large strains, the representation of mechanical response is proposed in engineering parameters (Figure II-13). The reason is that the main objective of these experiments is to verify the impact of drying temperature on the shear stress and shear strain without identification of material properties.

From the acquired data (Figure II-13), one can see that the mechanical response of different specimens is consistent despite three different desorption temperatures. One can notice a natural dispersion of experimental results; however, no important impact of temperatures on mechanical parameters is noted. Hence, one can assume that the drying temperature of +90°C does not noticeably degrade mechanical properties, while it significantly accelerates desorption kinetics. Consequently, it is decided to adopt this temperature for desorption of the studied composite of 2 mm thickness. Desorption duration is chosen as 10 days for the protocol; however, the mass stabilises within 8 days.

II.2.3. Development of humid ageing procedure for PA6 GF

Standard environmental conditions for automobile applications, as RH 50% and 85%, also require a protocol defining duration and temperature for the humid absorption process. To achieve this, specimens, dried at +90°C, are placed into a climatic chamber with controlled temperature and RH (Binder KMF-115, Figure II-14) to monitor the mass changes. The same gravimetric method as in Eq. 6 is used here for the data acquisition.

At first, a few specimens are exposed to RH 50% and two temperatures: $+50^{\circ}$ C and $+90^{\circ}$ C. The aim is to compare the absorption kinetics at two different thermal loadings. To recall, at RH 0% temperatures would be below and above T_g ($T_g \approx +23^{\circ}$ C for RH 50%) respectively, but once RH 50% is reached, both absorption temperatures are considerably above T_g .



Figure II-14: Climatic chamber Binder KMF-115 with controlled temperature and humidity

Humid absorption at $+50^{\circ}$ C and RH 50% lasted for 54 days and 28 days at $+90^{\circ}$ C and RH 50% (Figure II-15). It is important to mention that the saturation does not reach the equilibrium value at $+50^{\circ}$ C. In the case of $+90^{\circ}$ C, the maximum moisture content is reached in 7 days. Comparable results are presented by **[46,52]** for neat polyamides. As suggested by Taktak et al, Vlasveld et al. **[49,52]**, the diffusion coefficient increases with the ageing temperature and the latter stimulates the water uptake by the matrix of the composite. Such behaviour can occur due to the increased polymer chain motion in the amorphous part of the matrix, which is in a viscous state, thus, facilitating the penetration of water molecules with the consecutive hydrogen bonding to -NH- or carbonyl groups. The water uptake by the neat PA6 at the conditions $+25^{\circ}$ C/in water and $+24^{\circ}$ C/RH 80%, investigated by **[45,58]** respectively, reached the saturation state in nearly 200 days. The material T_g is thus still below the testing temperature at saturation. Therefore, in order to keep the consistency of desorption/absorption kinetics and to accelerate the procedure, also suggested by **[70]**, we select $+90^{\circ}$ C for the further ageing tests.

Analysing Figure II-15, one can notice the stabilised moisture content of +90°C/RH 50% within a relatively short period; nevertheless, the absorbed moisture tends to a slight decrease if the composite is exposed to a humid and hot environment for a long time period. Analogous behaviour is shown for another test batch at the same conditions (Figure II-24). It is presumed that the exposure of the studied composite or its matrix to elevated temperatures at constant RH for such prolonged period (28 days) can result in a mass loss that might signify the beginning of the chemical damage.

It is also necessary to estimate the absorption time for RH 85% at the chosen +90°C. One can see from Figure II-16, the increase of moisture contents as a function of time. For RH 85% the mass is stabilised in 2.5 days, and C_{glob} remains around 1.815%. Similar results are reported by Ishisaka et al. [55] for moisture

absorption of PA6 pellets at the constant temperature of $+50^{\circ}$ C. Regarding the relation between RH and water content of the composite PA6 GF, it is in good agreement to the experimental results of **[49,58,67]** for the matrix PA6. In order to ensure full saturation in moisture at RH 85% without causing chemical damage, 7 days are preferred for the protocol. It is then used for the saturation of specimens dedicated to the tensile testing campaign.

As presented in Figure II-16 and by Ishisaka et al. **[55]**, the absorption rate is higher for greater RH level. The initial increase of the moisture uptake is linear with the subsequent stabilisation and, thus, can be characterised by Fickian diffusion equation, as also shown by **[45]**. The results and suggestions of non-Fickian behaviour, proposed by **[21]** for PA6, are not in correlation with the given composite according to Figure II-16 and Figure II-21 in terms of non-increasing moisture content after the stabilisation; however, the smooth slope at the onset of saturation reports the opposite.



Figure II-15: Comparison of absorption by PA6 GF at the temperatures +50°C and +90°C



Figure II-16: Moisture absorption of PA6 GF at 90°C and RH 50%/RH 85%
The summary of humid conditioning for the composite PA6 GF is presented in Table II-1. The duration is selected as such that ensures the full desorption and absorption at given RH conditions. These conditioning parameters are applied to different geometries of PA6 GF for the final validation and repetitive humid ageing/desiccation in section II.2.4.

Condi	tioning parameters	С _{glob} , %	Duration		
Desorption:	+90°C, under vacuum	≈0	10 days		
Absorption:	+90°C, RH 50%	0.799	8 days		
	+90°C, RH 85%	1.812	7 days		

Table II-1: Conditioning of woven PA6 GF composite specimens

II.2.4. Protocol validation: case of neat matrix PA6 and circular PA6 GF samples

A verification procedure of defined desorption/absorption methods is realised in order to validate the protocol and simultaneously prepare specimens for testing campaigns. The physical reversibility of the composite is briefly reviewed during the repetitive humid ageing at RH 85%, followed by desorption.

Neat matrix PA6

The aim for conditioning the matrix specimens is, firstly, to verify whether the protocol parameters, defined for the composite, are appropriate for the neat matrix; secondly, to compare the manufactured neat matrix specimens to the matrix within the composite that underwent numerous processing stages; and, thirdly, to prepare neat matrix specimens for the mechanical experimental campaign. All the parameters, proposed in Table II-1, are maintained except the duration which is expected to become changed due to the different thickness, the absence of reinforcement and another storage condition. The effect of specimen geometry is investigated using three specimen types (Figure II-1 – Figure II-3).



Figure II-17: Desorption of neat matrix samples. Average of 10 specimens for batch 1, of 6 specimens for batches 2 and 3

Condi	itioning parameters	С _{дlob} , %	Duration		
Desorption:	+90°C, under vacuum	≈ 0	20 days		
Absorption:	+90°C, RH 50%	2.518	8 days		
	+90°C, RH 85%	5.502	7 days		

Table II-2: Conditioning of neat PA6 matrix specimens

The moisture content of specimens is unknown due to the uncontrolled storage; thus, they are subjected to the desiccation at first. The loss of moisture content over time is demonstrated in Figure II-17 for the dog-bone specimen type (Figure II-1). We can notice the typical mass reduction and stabilisation similar to the composite. The duration of desorption is, therefore, longer and the stabilisation occurs in 20 days for the batches 2 and 3 and 25 days for the batch 1.

In order to characterise the moisture absorption capacity of the matrix, the mass of dog-bone specimens is recorded for RH 50% and RH 85% and demonstrated in Figure II-18. During moisture uptake, the polymer seems to have a very similar tendency to behave as the composite PA6 GF. One can notice a slight decrease in C_{glob} over time for both RH. It is also interesting to mention that the geometry of specimens does not impact the absorption kinetics, as demonstrated in Figure II-19. The final conditioning parameters for the matrix are summarised in Table II-2.



Figure II-18: Humid ageing of neat matrix samples of type 1. Average of 10 specimens for RH 85% and average of 6 specimens for RH 50%



Figure II-19: Moisture uptake by the matrix as a function of specimen geometry

Nevertheless, the moisture contents of the matrix are visually in a good agreement with the moisture contents of the composite (Figure II-16 and Figure II-19). In order to confirm the moisture absorption by the composite matrix and to compare it to the moisture uptake by the matrix, some simple computations, based on volume and mass fractions of composite constituents, are necessary. The matrix content in the composite is 53% by volume [163]. If one considers the standard 45°-oriented specimen (Figure II-5) of volume V_{CM} and density ρ_{CM} , its mass is:

$$m_{CM} = \rho_{CM} V_{CM} \tag{10}$$

Hence, the volume of the matrix is:

$$V_m = V_{CM} \vartheta_{m \ volume} \tag{11}$$

The mass of the matrix is computed with Eq. 10 using only the matrix volume content and density. The matrix content by mass within the composite is the simple division of the matrix mass by the total composite mass (Eq. 12). Computed matrix properties are summarised in Table II-3.

$$\vartheta_{m\,mass} = m_m / m_{CM} \tag{12}$$

The mass fraction of the matrix within the composite is 33.56%. Supposing the matrix mass fraction of 100%, it would contain 5.399% of moisture at RH 85% and 2.381% at RH 50% according to the data in Table II-1. Comparing these results to the absorption by the neat matrix (Table II-2) at the same duration, the difference is 1.92% for RH 85% and 5.44% for RH 50%. The computed results demonstrate the similarity between the neat PA6 and the matrix within the composite, which have the common tendency to moisture absorption. The variation of 1.92% and 5.44% may arise from the presence of fibres restricting the movement of water molecules within the composite structure as cited in section I.1.4.2.

Property	Notation	Value	Measure unit
Density of PA6	$ ho_m$	1.14	g/cm ³
Density of PA6 GF	$ ho_{CM}$	1.80	g/cm ³
Mass of PA6	m_m	7.55	g
Mass of PA6 GF	m_{CM}	22.50	g
Matrix content by volume	$\vartheta_{m \ volume}$	53.00	%
Matrix content by mass	$artheta_{mmass}$	33.56	0⁄0
Volume of composite	V _{CM}	12.50	cm ³
Volume of matrix	V _m	6.63	cm ³

Table II-3: Matrix contents, density, mass and volume within the composite. Some properties are from [163]

Different geometries of PA6 GF

0° and 90°-fibre oriented composite

Specimens, presented in Figure II-20, are cut from the same laminate; thus, the initial state is identical. However, the loss of moisture content is similar for 0° and 90° orientations after 21 days of desiccation, but exceeds the loss of 45°-oriented specimens by 21.5%. Furthermore, while moisture absorption is stabilised (Figure II-21), the moisture content is higher in 0° and 90°-fibre oriented specimens for 7.16% as compared to 45°. Since the thickness of all the specimens remains the same of 2 mm, such differences may come from the geometry, stacking sequence and fibre orientation as also suggested by Jedidi **[25]** and Beringhier **[169]** for the carbon-epoxy composite material. Nevertheless, the stabilisation occurs within 8 days of exposure to 50% of RH and 10 days of desiccation for all stacking sequences.



Figure II-20 Moisture desorption of 0°, 90° and 45°-fibre oriented composites at RH 50% and 90°C



Figure II-21 Moisture absorption of 0°, 90° and 45°-fibre oriented composites at RH 50% and 90°C

Circular composite specimens

As mentioned in section II.1.2.2, the choice of circular specimens and their geometry (Figure II-9) is based on the numerical and experimental characterisation of prestressed bolted joints in Chapter IV, where the circular composite specimens are used as clamped parts. Hence the validation of desorption – absorption equally serves to verify the conditioning duration of different thicknesses prior to testing. In order to ensure the repeatability and consistency of results, 3 specimens are used for each thickness and diameter. They are initially subjected to desorption and then absorption at selected conditions in Table II-1.

The samples of 2 mm and 4 mm have coherent desorption kinetics, whereas its variation for 6 mm can be explained by the different initial state of the composite laminate due to the manufacturing 2.5 months prior to conditioning.



Figure II-22: Loss of moisture as a function of the square root of time divided by the specimen thickness

The results of humid ageing at RH 85% are introduced in Figure II-23 as a function of ageing time divided by the specimen thickness (Appendix A: Figure VII-4 for RH 50%). Therefore, all the specimens have similar final moisture content. When specimens are large and long enough, the penetration of molecules through the sides can be neglected. For circular samples of 2 mm (Appendix A: Figure VII-2) and rectangular samples of 250 × 25 × 2 mm (Figure II-16) the absorption time remains the same, supposing that in the case of samples with the diameters 18 mm and 36 mm the penetration of water molecules also occurs through the thickness, neglecting the sides. It explains the longer duration of absorption of thicker specimens (Appendix A: Figure VII-2) and Figure VII-3), for instance, specimens of 6 mm require at least 17 days for absorption, 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_{alob} is reached faster. This was also reported by Obeid [4].



Figure II-23: Moisture content as a function of the square root of time divided by the specimen thickness

Repetitive humid ageing – desiccation: RH 0% – 85% – 0%

Physical reversibility of PA6 GF is an important criterion for applications. The theory says that if a saturated specimen is subjected to a dry environment, it should recover to the DAM state if the material is reversible in terms of moisture uptake **[15,56]**. Then, during the subsequent exposure to the humid conditions, it should return to its saturated state. Hence, the objective of this test is to confirm or reject the reversibility of the composite material.

The environmental condition of RH 85% and +90°C is chosen for the cyclic absorption – desorption as the most severe. Three tested specimens are initially dried and stored into a sealing bag. Desorption and absorption procedures are completed according to the above-described protocol.

Specimens are initially subjected to drying at 90°C during 10 days. Each cycle consists of absorption at RH 85%/+90°C during 7-8 days with the consequent desorption at 90°C in vacuum during 10 days. As we can see from Figure II-24, C_{glob} at saturation is equal to that in Figure II-16 at the same conditions. However, the maximum moisture content slightly decreases with each next cycle: 1.764%, 1.728% and 1.711% respectively, thus 5.7% of decrease from 1st to 4th cycle. Regarding the minimum value of C_{glob} , it does not remain stable either: -0.058% at the end of the 1st cycle, -0.051% after the 2nd and -0.076% after the 3rd as compared to the initial dry state. The total decrease from the 1st to the 3rd cycle is 33%. The statement of fully reversible physical properties is not appropriate for PA6 GF unless the mechanical properties are tested. Evernden et al. [56], Grammatikos et al. [87] report similar mass loss during the hygro-thermal ageing. Authors relate this behaviour to the chemical decomposition, resulting in the material draining, and the matrix cracking. The latter should increase the moisture absorption capacity, which, however, is not proved in Figure II-24.

It should be noted that the degradation of mechanical properties with the increasing number of absorption – desorption cycles can also be induced. It is therefore revealed by Evernden et al. **[56]** that the shear modulus at DAM state remains reversible at the ageing temperature of +40°C. It is thus of great interest to thoroughly investigate mechanical behaviour at each cycle of desorption and absorption in terms of perspectives.



Figure II-24 Cyclic absorption – desorption: RH 0% ↔ RH 85%

Summary

The developed conditioning protocol is aimed to provide the necessary state of the composite PA6 GF. The known initial material moisture content would ensure an accurate mechanical response during experimental testing. However, the duration of desiccation and humid ageing varies for different composite material

thicknesses. Hence, a thick specimen requires more time to get the stabilised moisture content as compared to a thin specimen; and this duration is not proportional. Likewise, desorption may need more time. Although three thicknesses and several specimen geometries are tested in this work, the obtained results are not exhaustive. It is worth mentioning that different types of matrix coupons eventually provide the same values of moisture uptake. This, however, may not be the case with a composite due to the different fibre contents and the reinforcement types. It may be interesting to extend the present study for other kinds of composite structures and various contents of reinforcement. Furthermore, continuous conditioning cycles of "desiccation – humid ageing" reveal the minor losses of composite specimen mass after each desorption. It is supposed to be related to the chemical degradation, yet it necessitates more detailed experimental, chemical and mechanical analysis of the composite as well as the matrix.

The other important point consists in the obligation to control the environment during any mechanical testing, especially at high temperatures. This is strongly recommended due to the very rapid moisture loss during the first couple of hours of exposure to high temperatures as demonstrated for the composite PA6 GF and the matrix PA6 during the desiccation procedure. It is particularly important for great moisture contents.

Finally, it is proposed to readapt the developed conditioning protocol to the less sensitive to moisture composite material PPS CF. Although it is not expected to be relevant for the definition of elastic properties, the non-linear behaviour can be affected.

II.2.5. Protocol readjustment for PPS CF

According to the literature review on the composite material PPS CF (section I.1.5), its moisture absorption capacity is indeed unimportant for elastic properties. Nevertheless, a rigorous material characterisation necessitates an accurately predicted initial material state. It is also demonstrated in the literature review that the absorbed water molecules may cause material plasticisation, which can be important for damage initiation or elastoplastic behaviour. For this purpose, it is proposed to readapt the developed protocol for this composite. To apply an appropriate and accelerated conditioning to the present thermoplastic composite material, the formerly developed protocol can be adopted with some modifications. Hence, the desiccation and mechanical testing of DAM specimens are also followed by the absorption at RH 50% and RH 85%.

II.2.5.1. Desiccation of PPS CF

According to the technical data sheet of the carbon-reinforced PPS material, its glass transition temperature T_g is +90°C in the dry state. To accelerate desorption, the material must be subjected to a temperature above the ambient one and below T_g for a period of time. It should be reminded that the use of high temperature during sorption or desorption could lead to a change in the behaviour of the material. In order to evaluate the influence of drying temperatures on desorption kinetics and mechanical properties, we selected three temperatures close to T_g of the PPS CF: +70°C, +90°C and +120°C. For more significant desorption acceleration, all specimens are placed in the thermal chamber under vacuum (Figure II-10). The water content is deduced from a gravimetric analysis (Eq. 6) using a high-precision balance of 0.1 mg. In order to maintain the consistency of the results and avoid probable errors during analysis, a minimum of three specimens is conditioned for a given temperature.

The material state after desorption should be close to DAM, or RH 0%. The comparison of three desorption temperatures (Figure II-25) shows similar kinetics for +90°C and +120°C, while we observe slightly different kinetics at +70°C. After one week, the rate of moisture loss is almost equal for temperatures higher or equal to T_g . In addition, prolonged desorption at +90°C does not show any additional deterioration over time due to the absence of sudden mass loss or increase. It should also be noted that by reducing the temperature for desorption (+70°C) it is necessary to considerably increase desorption time since a certain water content is always present in the specimens [4]. This is also clearly visible on the graphic for C_{glob} at +70°C. Following the desorption



procedure, it can be seen that the composite, initially manufactured and conditioned in the open air, contains about 0.065% of moisture. All the specimens are placed into sealed bags before the further mechanical tests.

Figure II-25: Desorption of PPS CF as a function of temperature

In order to verify the degradation of mechanical properties resulting from desorption, we carried out tensile tests on each batch of specimens at room temperature (+23°C) (Figure II-26). Some variations in modulus and shear strain are observed. However, in general, no critical dependence on the conditioning temperature is visible, except in the non-elastic range, which is not of interest in the present research work. For more detailed analysis, further studies should be performed due to the significant variations of stresses and strains within the plastic range. Nevertheless, carried experiments allow us to select +90°C as desorption and, also, absorption temperature. Although under this temperature, desorption protocol requires one week, it is interesting to mention that the mass stabilisation occurs within two days.



Figure II-26: Mechanical behaviour of the dry composite under shear loading. Influence of the desorption temperatures: $+70^{\circ}$ C, $+90^{\circ}$ C and $+120^{\circ}$ C

II.2.5.2. Humid ageing of PPS CF



Figure II-27: Humid ageing of woven PPS CF as a function of RH

Specimens, dried at +90°C, are exposed to a controlled humidity and temperature environment (Binder KMF-115: Figure II-14) to monitor the absorption rate at RH 85% and 50%. The temperature of +90°C is also the maximum admissible by the climatic chamber (Figure II-14).

The results of the experimental wet ageing campaign (Figure II-27) enable determining the absorption kinetics of the composite material at RH 50% and RH 85%. First of all, there is some dispersion of the experimental results. It can be explained by susceptible weight measurements due to low moisture uptake. The moisture content is higher at RH 85% and equal to 0.0937%, which is close to the results obtained in the literature **[26,109]**. It takes about 8 days at RH 50%/+90°C and 16 days at HR 85%/+90°C (Table II-4) to obtain the stabilised moisture content. The proposed protocol can be adopted by researches prior to the rigorous characterisation of elastoplastic properties of this material; however, it may not be necessarily relevant to condition the composite for the determination of elastic characteristics due to indeed low water uptake.

These parameters are summarised in Table II-4 and then used to prepare the specimens for the experimental campaign of in-plane mechanical characterisation (section II.4).

Condit	ioning parameters	С _{дlob} , %	Duration, days		
Desorption:	+90°C, under vacuum	≈ 0	7		
Absorption:	+90°C, RH 50%	0.0384	8		
	+90°C, RH 85%	0.0937	16		

Table II-4: Conditioning of woven PPS CF composite specimens

II.2.6. Evaluation of glass transition temperature of PA6 GF as a function of moisture content

Sensitivity to moisture results in significant changes in the amorphous part of the matrix PA6, as assumed by [67,78]. The absorbed water molecules, acting as a plasticiser, lead to the reptation of molecular chains, similarly to the thermal effect. As outlined in section I.1.3.2, such behaviour contributes to reducing the glass transition temperature T_q , which is important to analyse for a rigorous material characterisation.

Regarding the composite material PPS CF, no crucial changes of T_g are revealed from the literature review. The most significant rise of the glass transition temperature in comparison to +90°C [164] is demonstrated by [109]. It is, therefore, supposed to be related to the post-curing. The humid ageing results in a change of only 1°C. Consequently, it does not represent any interest for the present work.

The range of T_g is evaluated with DMA, illustrated in Figure II-28. The moisture evaporation during these tests occurs due to high temperatures and represents a major challenge. In order to restrain it, the specimen's geometry is of $60 \times 10 \times 2$ mm dimensions; therefore, the moisture loss is negligible with respect to test duration. The analysis is conducted for initially conditioned material to ensure three moisture contents: RH 0%, 50% and 85%. Specimens are tested in a bending load mode. The correspondence to the onset of the drop in the storage modulus is used to define the critical value of T_g .



Figure II-28: Dynamic Mechanical Analyser

Resulting from the moisture impact, T_g alters from a positive to a negative temperature. According to DMA, the glass transition temperature is about +55.8°C in DAM state (Appendix C), which is coherent to +60°C as mentioned by the supplier [163]. When the full saturation occurs at RH 85%, T_g drastically decreases to a negative temperature, which is -1°C in our case. In the case of the standard atmospheric conditions, RH 50%, T_g is near the ambient temperature +23°C. Similar results are also shown by numerous authors [47,55,58,74,77,96] for filled and neat PA6. The origin of such a drop of T_g is interpreted in various articles. The plasticisation effect of moisture is the main contribution resulting in the intense motion of polymer chains.

	RH 0%	RH 50%	RH 85%	Type of analysis		Specimen
T _g	+55.8°C	+23°C	-1°C	DMA	Bending mode	$60 \times 10 \times 2 \text{ mm}$

Table II-5: Glass transition temperature for different moisture contents

II.2.7. Conclusions

The tendency of studied composite materials and neat matrix to moisture absorption is an essential factor that is to be taken into consideration before structure dimensioning. Each of three materials reacts to the presence of humidity in a different manner that depends on the material thickness, presence of reinforcement and its content, type of matrix, presence of flaws and their quantity within the material, which can be increased due to solicitations, type of environment, etc.

A protocol is necessary to ensure a condition of a composite material or a matrix according to required levels of relative humidity. As in the case of the present materials, such information is usually not provided by manufacturers; thus, it is not available in the open access. It has therefore been decided to develop a conditioning protocol for three studied materials – hygroscopic PA6 GF and PA6 and less sensitive to moisture PPS CF – to demonstrate the industrial partner how to characterise a material, sensitive to environmental conditions, in a reliable manner. The choice of both first materials is justified by high humidity impact, which causes essential changes in the glass transition temperature (from around $+60^{\circ}$ C in a dry state to -1° C when

saturated at RH 85%) and the mechanical behaviour. The proposition of conditioning another composite PPS CF is principally based on the fact that a rigorously evaluated material state at given RHs may demonstrate some variations in the elastoplastic behaviour, which is one of widely studied subjects in research. Hence, the conditioning methodology is likewise proposed for this material despite the principal interest in elastic properties in this research work.

The duration of material characterisation and subsequent validation of selected parameters are nearly eight months in total. The development is launched for the composite PA6 GF due to its hydrophilic behaviour and its main interest for the project. The selected desiccation and humid ageing temperature is $+90^{\circ}$ C in order to keep the consistency of desorption and absorption kinetics. This temperature is indeed above the material T_g in a dry state that theoretically may not be favourable for the chemical, physical and mechanical behaviour, but the mechanical testing of 45° -fibre oriented specimens did not show any critical changes for three tested temperatures. Nevertheless, the real working conditions for this composite are above its T_g , which may result in more critical degradation of properties in comparison with short-term exposure during conditioning. And, in addition, the chosen temperature greatly accelerates the process. Subsequently, this methodology is validated for other fibre orientations and thicknesses of the composite and the matrix PA6. Three specimen types of matrix demonstrated practically identical moisture contents at a given RH, which are consistent with the moisture content in PA6 GF. However, materials of different thicknesses do require the revision of duration of conditioning, which is, in addition, non-proportional. This aspect demands particular attention, and the duration should be restudied for each new thicknesse.

The rigorous material characterisation necessitates an accurate prediction of material state despite its low hygroscopicity. Hence, the developed methodology is readjusted for the sensitive composite material PPS CF. The same temperature of $+90^{\circ}$ C for desorption and absorption is selected through the mechanical testing of coupons dried at three temperature, different from those used for PA6 G. In addition, the maximum temperature for humid ageing is limited by the climatic chamber.

A brief investigation of PA6 GF material reversibility after repetitive humid ageing – desiccation demonstrated that the composite does not fully retain the initial mass when dried. However, mechanical behaviour after each cycle is not tested. It can demonstrate whether the properties could be recovered, which is of great interest in terms of perspectives for further material examination and exploitation. In addition, a more significant number of conditioning cycles may contribute to a better comprehension of the results. Also, it may be interesting to analyse the structure of such specimens for chemical or physical degradation.

II.3. ANALYSIS OF THERMAL EXPANSION AND HYGROSCOPIC SWELLING: IN-PLANE AND OUT-OF-PLANE CASES

The environmental conditions are usually represented in the form of thermal and humid impacts. If not accounted for during designing, thermal expansion (shrinkage) and/or hygroscopic swelling may cause the stresses inside the structure due to fluctuations of temperature or humidity. The materials, presented in this Chapter, tend to expand under such conditions. In order to thoroughly examine the studied composite materials PA6 GF, PPS CF and the matrix PA6, it is proposed to measure the coefficients of thermal, from +20°C to +80°C, and hygroscopic expansion for RH 50% and 85%. Two methods are used: manual measurements with a micrometre and strain gauges. These coefficients can be, therefore, used for numerical modelling if required.

II.3.1. Description of applied methods

Hygro-thermo-mechanical behaviour can be characterised by the constitutive equation written as follows:

$$\sigma_{ij} = C_{ijkh} (\varepsilon_{hk} - \alpha_{ij} \Delta T_t - \beta_{ij}^H M_t)$$
⁽¹³⁾

where σ_{ij} is the stress tensor, C_{ijkh} is the stiffness tensor, ε_{hk} – the strain tensor, α_{ij} and β_{ij}^{H} are the tensors of thermal and hygroscopic expansions respectively [25]. Highly sensitive to temperatures and/or humid impact composite materials may undergo a considerable change in dimensions; besides, the internal hygroscopic or thermal stresses may arise. Resulting from the literature review (Chapter I) and the studied hygroscopic swelling of PA6 GF and PPS CF composites (section II.2), it is particularly important to determine their dilatation and swelling coefficients. Hence, two methods, used by Westphal [170] and Jedidi [25], are proposed. All the experiments are realised at Research Institute of Civil and Mechanical Engineering (GeM).

Thermal dilatation

The thermal impact on composite structures may lead to the internal stresses notably if the in-service temperatures are elevated that is the case with PPS CF composite material. The suggested following methodology consists in the determination of in-plane coefficients of thermal expansion along the longitudinal and transversal fibre orientations. It can be therefore interesting for the applications in bolted joints to compute the coefficient in the thickness direction of both composites. However, it is not studied in the present work.

Experimental facility

The thermal impact on the woven composite materials is analysed with the use of bidirectional strain gauges, enabling the computations in longitudinal and transversal specimen directions. The materials are subjected to temperatures from about $+20^{\circ}$ C to $+80^{\circ}$ C (Figure II-29) with an increment of 10° C every 30 min. The specimens serve for the computation of **C**oefficients of **T**hermal **E**xpansion (CTE). The limited time duration is necessary to ensure the homogeneous temperature through the specimens. Besides, prolonged exposure at high temperatures may lead to the desiccation resulting in a coupling between thermal and hygroscopic strains. The studied composite specimens are not conditioned. An aluminium coupon with a known coefficient of thermal expansion and one free strain gauge are used in order to evaluate the accuracy of strain gauge results. One thermocouple per specimen serves for the temperature control on the specimen surface, and one thermocouple controls the temperature in the thermal chamber. To summarise, the following equipment is used:

- 2 specimens of 0°-fibre orientation of PA6 GF and PPS CF;
- 1 specimen in aluminium alloy;
- 6 thermocouples of type K;
- 6 strain gauges HBM (three-wire quarter bridge configuration, 350 Ohm);

- 1 module for strain gauges;
- 1 module for thermocouples;
- 1 data recorder module.



Figure II-29: Experimental facility for thermal expansion experiment

Computational procedure

A method, used for the evaluation of the coefficients of thermal dilatation, is based on a methodology formulated in the thesis of Westphal **[170]**. The exposure of materials to varying temperatures leads to the appearance of thermal strains if other loading is not applied. It can be represented in the following form:

$$\begin{cases} \varepsilon_{xx}^{t}(T) \\ \varepsilon_{yy}^{t}(T) \\ \sqrt{2}\varepsilon_{xy}^{t}(T) \end{cases} = \Delta T_{t} \begin{cases} \alpha_{xx} \\ \alpha_{yy} \\ \alpha_{xy} \end{cases} = \begin{cases} \varepsilon_{xx}^{t}(t,T) \\ \varepsilon_{yy}^{t}(t,T) \\ \sqrt{2}\varepsilon_{xy}^{t}(t,T) \end{cases} - \begin{cases} \varepsilon_{xx}^{t}(t=0,T-T_{0}) \\ \varepsilon_{yy}^{t}(t=0,T-T_{0}) \\ \sqrt{2}\varepsilon_{xy}^{t}(t=0,T-T_{0}) \end{cases}$$
(14)

where ε_{xx}^t , ε_{yy}^t , $\sqrt{2}\varepsilon_{xy}^t$ and α_{xx} , α_{yy} , α_{xy} are the thermal strains and coefficients of thermal expansion along x axis, y axis and in shear respectively (in global coordinates); ΔT_t is the temperature variation, T and T_0 are current and initial temperatures, t is the time step.

According to Herakovich [171], the change of temperature does not cause the shear strains if the orthotropic composite material is analysed in local coordinates; hence, the shear component can be neglected:

$$\begin{aligned} \alpha_{xx} &= \alpha_{11} \\ \alpha_{yy} &= \alpha_{22} \\ \alpha_{xy} &= 0 \end{aligned} \tag{15}$$

As mentioned by Westphal [170], the total strain, indicated by gauges, is related to both installation conditions and surrounding temperature. The gauges, used in the present work, are installed under the ambient environmental conditions; hence, their thermal deformation corresponds to the difference between the deformation at the initial (T_0) and the current (T) temperatures as presented in Eq. 14. Besides, the measurements of strain gauges undergo the influence of dilatation of gauge grid under varying temperature. Usually the grid (or gauge) coefficient is provided by manufacturers, which is $0.5 \ 10^{-6} \ ^{\circ}C^{-1}$ for the employed gauges here. This value can be introduced in a module to compensate for the thermal effects directly and to measure only mechanical strains. Another method is a compensation for the thermal effect on the grid (Figure II-30) by using a formulation provided by a manufacturer for each type of strain gauges. For the strain gauges, employed in this work, it is represented in Eq. 16. Both methods are eventually adopted for the computations in order to select the one that assures the accuracy of results.

$$\varepsilon_G^t = -16.56 + 2.4T - 8.59 \cdot 10^{-2}T^2 + 3.49 \cdot 10^{-4}T^3 \tag{16}$$



Figure II-30: Temperature response of the strain gauge 1-XY36-6/350 of the trademark HBM®

The third method, suggested by Westphal [170], consists in the compensation of thermal strain of grids with the use of a material with a known CTE. Thus, a specimen of aluminium alloy with $\alpha_{alu} = 23.8 \ 10^{-6} \ ^{\circ}C^{-1}$ between +20°C and +100°C is used. The deviation of strain data due to thermal impact $k_{T_1 \rightarrow T_2}$ can be detected by comparing total registered deformation to analytical [170]:

$$\varepsilon_{alu}^t = \alpha_{alu T_1 \to T_2} (T_2 - T_1) \tag{17}$$

$$k_{T_1 \to T_2} = \varepsilon_{alu}^t(T_2) - \varepsilon_{alu}^t(T_1) - \alpha_{alu \, T_1 \to T_2}(T_2 - T_1)$$
(18)

where ε_{alu}^t and $\alpha_{alu T_1 \to T_2}$ are the thermal deformation and CTE of the metal respectively in the range of temperatures $T_2 - T_1$; $k_{T_1 \to T_2}$ describes the strain data deviation between T_2 and T_1 , $\varepsilon_{alu}^t(T_2)$ and $\varepsilon_{alu}^t(T_1)$ are the total deformations of gauge at T_2 and T_1 respectively. Consequently, the coefficients of thermal expansion of the composites can be computed by eliminating the deformation of the gauge grid [170]:

$$\alpha_{11\,T_1 \to T_2} = \frac{\varepsilon_{xx}^t(T_2) - \varepsilon_{xx}^t(T_1) - k_{T_1 \to T_2}}{T_2 - T_1} \tag{19}$$

$$\alpha_{22\,T_1 \to T_2} = \frac{\varepsilon_{yy}^t(T_2) - \varepsilon_{yy}^t(T_1) - k_{T_1 \to T_2}}{T_2 - T_1} \tag{20}$$

The results of the coefficient of thermal expansion in transversal and longitudinal fibre orientations are presented and discussed in section II.3.2. for both composite materials.

Hygroscopic swelling

The hygroscopic expansion of material is the deformation ε_H resulting from the variation of mass M_t due to the moisture uptake at a given temperature. The coefficient of hygroscopic expansion noted as β_H , is not widely studied due to the complexity of experimental testing; thus, a few experimental results are found [25,45]. The absorption of moisture during in-service life may result in the problematic, similar to the thermal impact, in particular, in the dimensional increase. Hence, it is proposed to estimate the properties of the composite PA6 GF and the matrix PA6 in through-thickness direction. The composite material PPS CF is not studied due to small moisture uptake that makes it challenging to analyse the out-of-plane swelling accurately. The out-of-plane direction is selected for its interest in the design of bolted joints.

$$\varepsilon^H = \beta^H_{ij} M_t \tag{21}$$

The matrix and composite specimens are initially dried in order to eliminate the moisture. Then they are subjected to the environment that corresponds to the developed conditioning protocols. Hence, the thermal impact is present. The same methodology as for conditioning is used here. Therefore, in addition to mass, the thickness is also measured with a micrometre. Three measurements per specimen are taken for the reproducibility. The results for RH 50% and RH 85% are presented in section II.3.3. The other approach using strain gauges is suggested by Jedidi [25]; however, it is not employed in the present work.

II.3.2. Results of thermal expansion

Figure II-31 and Figure II-32 present the evolution of the thermal in-plane strain of the aluminium coupon and its coefficient of thermal expansion in longitudinal and transversal orientations. The type of used gauges enables the computation of CTE in both directions; thus, it confirms their proper functioning due to the equality of obtained results (Figure II-31) despite the used method. Nevertheless, the use of the temperature compensating equation (Eq. 16) seems more appropriate for the following computations than the application of gauge coefficient. The employed strain gauges are specific for the applications with composite materials, which may be a reason for such a difference between results. Supposing that the CTE of the aluminium coupon is $23.8 \ 10^{-6} \ \circ C^{-1}$, the thermal compensation equation represents well gauge behaviour. Hence, this equation is consequently applied to the composite materials in order to compare the results to the approach, proposed by [170] and introduced in the previous section.



coupon

PA6 GF

The obtained results of thermal strains for PA6 GF composite material are presented for both approaches. Thus, Figure II-33 describes the in-plane strain of two specimens in two fibre directions. The compensating strain ε_G^t is integrated in Eq. 19 and Eq. 20 instead of $k_{T_1 \to T_2}$ coefficient. The results in Figure II-34 are entirely based on the approach, demonstrated in Eq. 17 – Eq. 20.





Figure II-33: Thermal strains ε_{xx}^t of PA6 GF in longitudinal and transversal fibre orientations with the use of ε_G^t

Figure II-34: Thermal strains \mathcal{E}_{XX}^t of PA6 GF in longitudinal and transversal fibre orientations with the use of $k_{T_1 \rightarrow T_2}$

The coefficients of thermal expansion along and transversal to fibres are outlined in Table II-6 for temperatures between +25°C and +80°C. The similarity of longitudinal and transversal coefficients is explained

Coefficien expa	t of thermal Insion	Specimen 1	Specimen 2	Average
c aT	$\alpha_1 (10^{-6} {}^{\circ}\mathrm{C}^{-1})$	10.449 ± 2.476	6.575 ± 0.832	8.512 ± 2.476
IOT EG	$\alpha_2 (10^{-6} \circ C^{-1})$	9.015 ± 2.039	9.450 ± 2.014	9.233 ± 2.039
for $k_{T_1 \to T_2}$	$\alpha_1 (10^{-6} {}^{\circ}\mathrm{C}^{-1})$	10.389 ± 5.027	9.298 ± 4.260	9.844 ± 5.027
	$\alpha_2 (10^{-6} \circ C^{-1})$	6.965 ± 4.532	9.726 ± 4.323	8.346 ± 4.532

by the balanced orthotropic woven structure of the reinforcement. Generally speaking, the dimensions of obtained values are coherent; however, the dispersion of results is present.

Table II-6: Coefficients of linear thermal expansion of PA6 GF

PPS CF

The same results are likewise extracted for PPS CF composite material. The thermal strain evolution is illustrated in Figure II-35 and Figure II-36. One can note that a deviation occurs at +70°C and +80°C. It is supposed to be related to the reused aluminium coupon, which is used the second time; thus, the adhesive deterioration could take place. The composite structure is likewise orthotropic and balanced that justifies the correspondence of properties.





Figure II-35: Thermal strains ε_{xx}^t of PPS CF in longitudinal and transversal fibre orientations with the use of ε_G^t

Figure II-36: Thermal strains \mathcal{E}_{xx}^t of PPS CF in longitudinal and transversal fibre orientations with the use of deviation $k_{T_1 \rightarrow T_2}$

Coefficient of thermal expansion		Specimen 1	Specimen 2	Average
í at	$\alpha_1 (10^{-6} {}^{\circ}\mathrm{C}^{-1})$	3.018 ± 0.351	3.445 ± 0.269	3.232 ± 0.351
for $\mathcal{E}_{\mathcal{G}}$	$\alpha_2 (10^{-6} \circ C^{-1})$	2.878 ± 0.246	3.112 ± 0.345	2.995 ± 0.345
for $k_{T_1 \to T_2}$	$\alpha_1 (10^{-6} \circ C^{-1})$	4.497 ± 1.946	4.717 ± 1.218	4.607 ± 1.946
	$\alpha_2 (10^{-6} \circ C^{-1})$	4.338 ± 1.858	4.709 ± 1.282	4.524 ± 1.858

Table II-7: Coefficients of linear thermal expansion of PPS CF

Summary

The principal idea of this small experimental campaign is to estimate the possible changes in material dimensions due to thermal impact. The computations are based on the specimen instrumentations with bidirectional strain gauges. The parameters are calculated by using two methods to compensate for the thermal grid expansions. These are the use of a reference aluminium specimen and the application of the equation to compensate for the thermal impact on the gauge itself. Obtained results seem promising for both composite materials; nevertheless, better precision remains necessary. One can note that the longitudinal and transversal coefficients and thermal strains are alike. It demonstrates that the thermal shear values are of practically zero

values, which is in agreement with the theory. However, the used composite coupons were not conditioned before the experiment; hence, there is a possibility that the desiccation process could take place at high temperatures. Such coupling can result in lower values of studied parameters. As a consequence, a more rigorous approach for the evaluation of CTE and thermal strains is required. A study of through-thickness properties may be interesting for the composite applications in bolted joints. The identified parameters serve for a general comprehension of material behaviour and are not employed in the following sections.

II.3.3. Results of hygroscopic swelling

PA6

Mentioned in the literature review, one of the results of hygroscopic impact is the dimensional changes. The expansion is associated with the amount of absorbed moisture as presented for the rectangular matrix specimens (Figure II-2 and Figure II-3) in Figure II-37 and Figure II-38. Thus, the exposure to high relative humidity naturally results in more significant hygroscopic deformation. Besides, the specimen geometry, thickness in particular, also presents an impact on the kinetics of moisture uptake. More graphs can be found in Appendix B.

From the figures below, one can notice a significant fluctuation of results. It is due to the data acquisition method. The precision of the micrometre is 0.001 mm, which appears not to be sufficient for the rigorous evaluation of thickness. Nevertheless, globally analysing Figure II-37, one can notice the stabilisation of out-of-plane strain in some time. Such behaviour is related to the saturation in the moisture of the matrix samples. Considering Figure II-38, the hygroscopic out-of-plane strain appears to be linearly related to the moisture content regardless of the specimen type and level of RH. The coefficient of hygroscopic expansion β_{ij}^{H} is represented by the slope of the linear relation. It is not computed in this work due to the scattering of results. For that, a more precise acquisition method is necessary.



Figure II-37: Out-of-plane hygroscopic deformation of matrix specimens at RH 50% and RH 85% over square root of time



Figure II-38: Out-of-plane hygroscopic deformation of matrix specimens at RH 50% and RH 85% versus moisture uptake

PA6 GF

The exact methodology, described above, is applied to the circular composite specimens, presented in Figure II-9 in section II.1.2.2. However, it is more complex to accurately measure the change in thickness due to the presence of reinforcement that restricts the hygroscopic expansion. Thus, the moderate level of relative humidity results in a significant data scattering comparing to RH 85%. It is aggravated by the small dimensions of specimens (Figure II-39). The specimens of small diameters are highly probable to undergo the penetration of water molecules not only through the thickness but also through the edges. Thus, the change of diameters should be evaluated as well. Besides, the heterogeneous structure does not facilitate the accurate measurements due to the small diameter of the spindle. Zones, rich in reinforcement or matrix, do not necessarily provide the

same result. Nevertheless, similarly to the matrix coupons, the out-of-plane hygroscopic strain seems to stabilise over some time, which is also related to the stabilisation of moisture uptake. For instance, the specimens of thickness 6 mm do not form horizontal lines since they may require more time for the saturation than the duration of the presented tests (Figure II-40). Some more graphs are demonstrated in Appendix B.



Figure II-39: Out-of-plane hygroscopic swelling of circular composite specimens of diameter 18 mm versus specimen thickness and RH



Figure II-40: Out-of-plane hygroscopic swelling of circular composite specimens of diameter 36 mm versus specimen thickness and RH

Summary

Although the use of the micrometre screw gauge does not provide very accurate results, the global tendencies of material behaviour can be estimated. The studied matrix PA6 and the composite PA6 GF demonstrate the relation of hygroscopic out-of-plane expansion to the moisture uptake ratio. Thus, the hygroscopic deformation at RH 50% is smaller in comparison with RH 85%. The relation between moisture uptake and hygroscopic strain is linear, which enables the computation of the hygroscopic expansion coefficient if the accuracy of data is improved. The low RH level leads to the important scattering of results due to the low precision of taken measurements, which is intensified by the heterogeneous structure of the composite and viscous state of the entire geometry of specimen should be considered if the overall dimensions are comparable with thickness. The moisture penetration should be controlled on the specimen sides. In terms of perspectives, a decoupling between thermal and humid impact may be of great interest for researches. What is more, the use of more precise experimental methods would provide a good database, if tested for different humid conditions, with a possibility to obtain the hygroscopic expansion coefficients. Nevertheless, the provided results are not adopted hereafter and mainly serve for the general material characterisation.

II.4. EXPERIMENTAL CAMPAIGN FOR IN-PLANE MECHANICAL CHARACTERISATION

The objective of the experimental campaign is to reveal modifications in mechanical properties of the composite materials and the matrix subjected to different climatic conditions. The influence of temperature and relative humidity is studied during mechanical tests. A complete study of mechanical properties is performed with the evaluation of elastic, ultimate and damage properties of each material as a function of testing conditions. However, the main focus of the research is on the elastic parameters; hence, the damage properties are not presented in the section, but are expected to serve for more detailed investigation of material behaviour afterwards. The selected environmental conditions, fibre orientations and data acquisition methods are described in the section. Resulting from the large strains, the mechanical properties are presented in "true" values.

II.4.1. Definition of test matrices

Knowing the impact of humid environment and working temperatures on the material behaviour of PA6 and PA6 GF (sections I.1.3 and I.1.4), it is essential to examine the influence of the biggest possible range of climatic conditions. In the case of PPS CF, the impact of high temperature on its properties is obvious (section I.1.5). The humidity impact is known to be low, except under the extremely humid environment (water immersion). Nevertheless, a proposed thorough mechanical characterisation includes dry and humid-aged specimen states. As mentioned in the previous sections, the elastic properties are not expected to be greatly affected, but the non-linear behaviour and damage properties may undergo some changes due to moisture uptake. This may be important for future research studies.

The environmental conditions should, therefore, correlate with the actual in-service environment that depends on the applications. Therefore, the testing conditions can be represented as a hygro-thermo-mechanical coupling (Figure II-41) due to the impact of climatic conditions and tensile forces.

All specimens are firstly referenced, then subjected to desorption and/or absorption procedures and, hence, directly placed into sealing storage bags. One of the strong assumptions, applied here, is homogeneously distributed moisture through the thickness of a specimen.



Figure II-41: Hygro-thermo-mechanical coupling be inspired by [42]. Domains of interest are highlighted in bold

PA6 GF and PA6

The standard temperatures for automobile industry, where the materials are widely employed, are from -40° C to $+80^{\circ}$ C and the common RHs are 0%, 50% and 85%. Thus, within the framework of this research, these environmental conditions are selected for PA6 GF due to the restricted available timespan, though they provide a rich database (Table II-8).

Present woven material is quasi-balanced according to the supplier; thus, the longitudinal (0°) and transverse (90°) properties should be similar; however, this statement is to be confirmed. Fibre properties are assumed to be independent of environmental conditions; thus, only one condition of RH 50% is applied **[172]**. All tests of 0° and 90°-oriented specimens are monotonic due to fibre dominated behaviour. There are four specimens of type 5 (Figure II-7) per each environmental conditions. Three specimens are loaded for the repeatability of results, whereas the fourth specimen is not loaded and serves as a "reference" one. It is used for the evaluation of the mass change during testing, which may occur at high temperatures due to specimen desiccation. This method consists in weighing specimens right after the bag opening, then placing it into the climatic chamber next to a loaded specimen and reweighing it when all tests are accomplished.

Specimens of 45°-oriented fibres exhibit in-plane pure shear behaviour under applied tensile forces. Thus, the mechanical properties are dominated by the matrix that is highly impacted by the environmental conditions. The loading rate might also cause some alteration of the material response, as in the case with the neat matrices PA6 and PA66 **[23,47]**. Either to reveal or to reject this information in the case of the composite material, it was decided to carry out extra experiments at 0.5 mm/min in the most severe among given conditions, whereas the rest of specimens is loaded at the rate of 2 mm/min. For the repeatability of results, we use six specimens of type 4 (Figure II-5) per condition, and the sixth one serves as the "reference specimen". Bigger number of coupons is used due to possible variation of results, highly dependent on the matrix response. The first test is monotonic to the failure, whereas the other four specimens undergo cyclic quasi-static loading to obtain the damage evolution.

As for the matrix PA6, its testing conditions fully correspond to Table II-8. Identic climatic conditions and velocity rates are necessary in order to accurately evaluate the similarity of the mechanical response of the matrix and of the composite, loaded in shear. In addition, obtained experimental properties of the matrix are used in Chapter III for numerical computations.

There are 4 specimens of type 1 (Figure II-1) per each environmental condition, where 1st and 2nd undergo monotonic loading, 3rd is subjected to cyclic quasi-static, and 4th is the reference specimen.

Temperature	RH 0%	RH 50%	RH 85%
-40°C	45°/R	45° 0° 90°/R	45°/R
-10°C	45°/R		45°/R
+23°C	45°/R	45° 0° 90°/R	45° 45°*/R R*
+40°C	45°/R		45° 45°*/R R*
+80°C	45° 45°*/R R*	45° 0° 90°/R	45° 45°*/R R*
		45°* 0°* 90°*/R*	

Table II-8: Test matrix for PA6 GF and PA6. 45° 0° 90° are the fibre orientations. R stands for the neat matrix. An asterisk stands for the loading rate of 0.5 mm/min; the rest is 2 mm/min

PPS CF

The present material is known to be used in applications, working in more severe environments than PA6 GF. It is thus essential to examine the impact of the temperature range for aeronautical applications (Table II-9). Negative temperatures are not examined. In the context of our research, two extreme values of relative humidity are applied: RH 0% and RH 85%. Despite the limited environmental conditions, the results supplement the experimental database of the present material.

The choice of 45°, 0° and 90°-oriented specimens is clarified in the section above. Similarly, the specimen types and their quantity are equal to PA6 GF with 1 reference specimen for every coupled condition "relative humidity – temperature". The solicitation mode is the tensile monotonic and cyclic quasi-static loading at 2 mm/min loading rate for all the specimens.

Temperature	RH 0%	RH 85%
+23°C	45° 0° 90°	45°
+90°C	45° 0° 90°	45°
+105°C	45° 0° 90°	45°
+120°C	45° 0° 90°	45°

Table II-9: Test matrix for PPS CF. 45° 0° 90° are the fibre orientations. The loading rate is 2 mm/min

II.4.2. Equipment and data acquisition

The experiments at positive temperatures are conducted at Research Institute of Civil and Mechanical Engineering (GeM) on the Instron 5584 testing machine, equipped with a load cell of 150 kN and a climatic chamber with temperature regulation (Figure II-44). For 0° and 90° specimens, data is acquired by means of strain gauges (Figure II-42 a) and images of effective length taken with a high-resolution camera Stemmer GT 6600 via VIC-SNAP software for DIC. To obtain the strain field of the specimen surface, each of them is speckled with white and black colour in order to obtain a speckle pattern on the effective length (Figure II-43). By reason of large deformation of 45° and neat matrix specimens, DIC measurement is mainly performed. For the verification purposes, a few specimens are instrumented with strain gauges (Figure II-42 b and c). The comparison of acquired results with strain gauges and DIC is performed and demonstrated in Appendix E.



Figure II-42: a) Instrumented biaxial strain gauge on 45°-fibre oriented specimen; b) biaxial strain gauges on 0°-fibre oriented specimen; c) instrumented axial strain gauge on neat matrix



Figure II-43: Dumbbell-shaped composite specimen with a speckle pattern



Figure II-44: Experimental set-up for tensile tests



Figure II-45: Biaxial clip-on extensometer

All the experiments at negative temperatures are carried out on the testing machine Instron 4505 with 100 kN load cell at CETIM Nantes. Due to the cold-gas circulation, taken images contain noise, thus longitudinal and transversal extensioneters are used (Figure II-45).

The reference specimen in each batch is neither instrumented nor speckled and serves for mass tracking before and after experiments. All tested and reference specimens are also instrumented with the thermocouples in order to monitor the testing temperature.

The data, acquired by means of the strain gauges, the extensioneters and the camera, is used for the computation of macroscopic properties of the composite material. Two commercial types of software (VIC 2D and TEMA) are used to process the results and to certify their accuracy. The first allows the simulation of a strain field, the use of numerical strain gauges, etc., while the latter consists of the point tracking and can show the specimen elongation, fibre reorientation etc. The results from VIC 2D and extensioneters are employed for the analysis of the mechanical state, whereas the fibre reorientation is characterised with TEMA.

II.4.3. Computational procedure

In this section, we propose a brief description of computational methods. Resulting from large strains, observed during tensile testing, it was decided to represent the results in "true" values rather than in "engineering" [173]. The data is retrieved from the recorded values of load cell and strain field, analysed in VIC 2D.

II.4.3.1. Composite materials

Unprocessed experimental data is obtained in global coordinates, also called "machine" frame. It is, however, more convenient to analyse composite mechanical properties in local coordinates that are defined by fibre orientations (Figure II-46). The transfer from the global frame to the local frame is related to the tensile direction of specimens and is achieved using the following equations [14]:



Figure II-46: Frames of elementary ply. Global coordinates in black $(0, \vec{b}_x, \vec{b}_y)$ represent the "machine" frame; local frame in red $(0, \vec{b}_1, \vec{b}_2)$ represents orthotropic coordinates [1]

$$\begin{pmatrix} \sigma_{11} \\ \sigma_{22} \\ \sqrt{2\sigma_{12}} \end{pmatrix} = P \begin{pmatrix} \sigma_{xx} \\ \sigma_{yy} \\ \sqrt{2\sigma_{xy}} \end{pmatrix}$$
(22)

$$\begin{pmatrix} \varepsilon_{11} \\ \varepsilon_{22} \\ \sqrt{\gamma_{12}} \end{pmatrix} = P\begin{pmatrix} \varepsilon_{xx} \\ \varepsilon_{yy} \\ \sqrt{\gamma_{xy}} \end{pmatrix}$$
(23)

$$\begin{cases} \gamma_{xy} = 2\varepsilon_{xy} \\ \gamma_{12} = 2\varepsilon_{12} \end{cases}$$
(24)

$$P = \begin{pmatrix} \cos^2\theta & \sin^2\theta & \sqrt{2\cos\theta\sin\theta} \\ \sin^2\theta & \cos^2\theta & -\sqrt{2\cos\theta\sin\theta} \\ -\sqrt{2\cos\theta\sin\theta} & \sqrt{2\cos\theta\sin\theta} & \sqrt{\cos^2\theta - \sin^2\theta} \end{pmatrix}$$
(25)

The change of frame and the use of specimens with three fibre orientations allow us to compute longitudinal, transversal and shear properties:

• Ultimate values of stresses ($\sigma_{11 max}$, $\sigma_{22 max}$, $\sigma_{12 max}$) and strains ($\varepsilon_{11 max}$, $\varepsilon_{22 max}$, $\gamma_{12 max}$);

• Elastic parameters as Young's modulus (E_{11} , E_{22}), shear modulus (G_{12}) and Poisson's ratios (ν_{12} , ν_{21}).

To achieve the calculation of the mentioned parameters, the step consists in compution of true strains (Eq. 28 - 29) and stresses (Eq. 30) in the global frame by using the "engineering" measured data (Eq. 26 - 27). Eq. 30 is valid for incompressible materials as well as for composites **[173]**. Though the computation of stresses is generally simplified by assuming that $-\dot{\epsilon}_{yy}^*/\dot{\epsilon}_{xx}^* = 1/2$ (Eq. 31) and is based on the assumption of material incompressibility, we take into consideration the contraction coefficient.

$$\varepsilon_{xx \ eng} = \frac{L - L_0}{L_0} \quad ; \quad \varepsilon_{yy \ eng} = \frac{W - W_0}{W_0} \tag{26}$$

$$\sigma_{xx\,eng} = \frac{F}{S_0} \tag{27}$$

$$\varepsilon_{xx\,true} = ln(1 + \varepsilon_{xx\,eng}) \tag{28}$$

$$\varepsilon_{yy\,true} = ln \big(1 + \varepsilon_{yy\,eng} \big) \tag{29}$$

$$\sigma_{xx\,true} = \sigma_{xx\,eng} \left(1 + \varepsilon_{xx\,eng}\right)^{2\nu^*} \tag{30}$$

$$\nu^* = -\dot{\varepsilon}_{\nu\nu}^* / \dot{\varepsilon}_{xx}^* \tag{31}$$

The second step consists in the computation of the quantities in the local frame by using the transformation matrix (Eq. 25) adapted to the required orientation of composite. In the case of the longitudinal and transverse direction, the value of the angle θ is equal to 0. Hence, the calculation of the longitudinal and transverse strains and stresses is straightforward (Eq. 32 – 33). Despite the small deformations and negligible effect on the final stress – strain data, longitudinal and transversal stresses and strains are likewise represented in "true" values in order to keep consistency of computations and results.

$$\theta = 0^{\circ} ; \quad \varepsilon_{11\,true} = \varepsilon_{xx\,true} ; \quad \varepsilon_{22\,true} = \varepsilon_{yy\,true} ; \quad \sigma_{11\,true} = \sigma_{xx\,true}$$
(32)
$$\theta = 90^{\circ} ; \quad \varepsilon_{11\,true} = \varepsilon_{yy\,true} ; \quad \varepsilon_{22\,true} = \varepsilon_{xx\,true} ; \quad \sigma_{22\,true} = \sigma_{xx\,true}$$
(33)

As for the in-plane shear properties, 45° -fibre orientation is a particular case for computations ($\theta = 45^{\circ}$). Resulting from the transformation to the local frame (Eq. 22 – 25), the shear properties are characterised with Eq. 34 - 35.

$$\gamma_{12 true} = \varepsilon_{xx true} - \varepsilon_{yy true} \tag{34}$$

$$\sigma_{12 true} = \frac{\sigma_{xx true}}{2} \tag{35}$$

Clarification of in-plane shear properties is illustrated on the theoretical shear stress – shear strain curve in Figure II-47. The shear and elastic moduli and the corresponding yield stress can be estimated with different methods. For instance, the authors in [174] report using the slope at 0.5% of a maximum shear strain for the estimation of a shear modulus. Other authors, [58], use the slope from 0% to 1% of strain for the computation of tensile modulus and up to 5% of deviation from the linear slope for the yield stress computation of the matrix PA6. However, no formally stated method is given for this type of calculations for composite materials. Therefore, we selected another more reproducible approach based on a simple linear regression, where a modulus and yield stress correspond to the maximum or fixed value of the coefficient of determination R^2 , calculated from the stress and strain experimental data (Eq. 36). Eventually, the maximum coefficient R^2 is adopted for the definition of elastic limit due to the rapid non-elastic behaviour of material under certain environmental conditions. This choice provides the consistency of all the results. Whatever the material and testing condition, the first five proposed points of shear and tensile moduli are not selected as maximum. This method ensures the identical data treatment for all acquired experimental results.

$$R^{2} = max(1 - \frac{\Sigma(y_{i} - f_{i})^{2}}{\Sigma(y_{i})^{2}})$$
(36)



Figure II-47: Theoretical shear stress - shear strain curve with the utilised notation

The same previous methodology is applied to identify the longitudinal and transverse properties.

II.4.3.2. Neat matrix

Resulting from the experimental campaign, we can evaluate isotropic matrix properties:

- Stress (σ_{max}) and strain (ε_{max}) ultimate values;
- Elastic parameters as Young's modulus (*E*), Poisson's ratio (ν) and yield stress (σ_{ν}).

The mechanical response is characterised with Eq. 37 - 38. As for the tensile modulus, the same approach as for the composites is applied (Eq. 36). The notation is proposed here as for the isotropic material; however, depending on a manufacturing method, the longitudinal, transversal and out-of-plane properties may vary.

$$\varepsilon_{xx \, true} = \ln(1 + \varepsilon_{xx \, eng}) \quad ; \quad \varepsilon_{yy \, true} = \ln(1 + \varepsilon_{yy \, eng}) \tag{37}$$

$$\sigma_{xx \, true} = \sigma_{xx \, eng} \left(1 + \varepsilon_{xx \, eng} \right)^{2\nu^*} = \frac{F}{S_0} \left(1 + \varepsilon_{xx \, eng} \right)^{2\nu^*} \tag{38}$$

II.4.4. Elastic and ultimate properties of PA6 GF

Longitudinal and transverse orientations

Woven laminate structure, with the identical type and number of fibres in warp and weft orientations, possesses theoretically equal longitudinal and transversal properties. Nonetheless, the actual mechanical response can slightly differ from 0° to 90°-fibre orientation as a result of a technological process. Experimental testing demonstrates the fibre-dominated properties of the laminate as well as their modest variations under hygro-thermal conditions.

Figure II-48 depicts the in-plane stress-strain results of longitudinally and transversally oriented specimens as a function of testing temperature. Hypothetically, the environmental temperature and the moisture content should not significantly impact the mechanical properties due to fibre domination. It is clearly visible in Figure II-48 that the elastic moduli remain practically unchanged at +23°C and +80°C, however both E_{11} and E_{22} increase for about 29% at -40°C. It may result from increased material stiffness when below T_g . All the values are summarised in Table II-11 at the end of the present section. A large scattering of tensile strength results is

caused by rupture occurring on the effective length of the specimens and near the grips. Nevertheless, it does not modify the estimation of Young's moduli. Comparison of warp and weft orientations demonstrates that the longitudinal properties are, however, slightly superior to the transversal. It can occur due to the weaving method when the transverse weft yarns are subjected to undulations in order to create a weave pattern, whereas the warp yarns remain fixed and in tension.

The tensile strain rate has a negligible effect on the longitudinal characteristics, less than 1% of the difference, while the transversal properties decrease by about 7% at low testing velocity (Table II-11).



Figure II-48: Stress-strain response of PA6 GF, oriented at 0° and 90°

Shear orientation



Characterisation of environmental impact

Figure II-49: Stress-strain response of PA6 GF at RH 0%

For the 45°-oriented specimens, their properties are dominated by PA6 matrix. Thus, the effect of the environmental conditions on the woven composite material can be demonstrated. The experimental results are summarised and reported in Table II-11.



Figure II-51: Stress-strain response of PA6 GF at RH 85%

The composite is tested at varying moisture contents and temperatures with the total number of the combinations equal to 15. The mechanical behaviour as a function of thermal loading is presented in Figure II-49 – Figure II-51, whereas the humidity levels are constant: 0%, 50% and 85% of RH correspondingly. Equally, the testing velocity remains 2 mm/min for every batch.

As one can see in Figure II-49 – Figure II-51, in-plane shear behaviour is significantly impacted by the testing temperature. The increase of the latter leads to the reduction of the shear strength and the partial growth of the shear strain at the same moisture content. The most important changes occur at $+23^{\circ}$ C and $+40^{\circ}$ C. At $+80^{\circ}$ C the drop in strength and the growth in strain take place with the rise of the moisture, however, at -40° C more

significant decrease of the mechanical properties is obtained in the current work as compared to 10% difference in **[86,175]**.

It is also worth mentioning that the shear strain at failure tends to decrease for RH 85% at elevated temperatures in comparison with RH 0% and RH 50%. It is supposed to occur beyond a critical value of moisture content as suggested by [47,52] for a neat matrix, noted as a "threshold effect". Images of tested samples are illustrated in Appendix D.

It is important to note that some curves are quite separated from the rest. This can be explained by the varying T_g that contributes to the changes in the physical state of specimens. It signifies that the amorphous part of the material is solid and fragile below this temperature, but becomes flexible and viscous if the surrounding temperature is above T_g . In addition to the testing temperature, one can also observe a decrease in T_g with the rise of the RH level (section II.2.6 Table II-5). Such a connection between the environmental temperature and RH provokes a coupled problem with regard to mechanical response. To summarise the aforementioned information, the brief conclusions are the following:

- In-service temperature and relative humidity have a strong impact on the in-plane shear behaviour;
- Decrease of shear strength, yield shear stress and shear modulus with rising temperature and RH;
- Growth of shear strain with rising temperature and RH;
- Considerable change of mechanical behaviour from rigid to viscous in the vicinity of T_q .

Figure II-52 illustrates the global state of the woven PA6 GF as a function of testing temperature and RH level. Hence, the importance of the environment as one of the major parameters characterising the composite material is demonstrated. One of such parameters can be introduced in the form of a temperature difference ΔT , calculated as the testing temperature T subtracted from T_a :

$$\Delta I = I_g - I \tag{39}$$



Figure II-52: Reassembled shear stress-strain curves of PA6 GF

The upper and the bottom graph parts of Figure II-52 are divided by the dashed black line matching the stress-strain curve with ΔT being practically zero. The upper half represents the solid state of specimens and the bottom one the viscous state, resulting from exceeding the defined T_g . Consequently, the PA6 GF material demonstrates a significant dependence on the working conditions.

(20)

Furthermore, certain mechanical responses may coincide or be relatively similar even though the specimens are of diverse environmental state. It can thus be explained by the adjacent values of ΔT for pairs "testing temperature – relative humidity". One can suggest that as long as ΔT is the same or close for a number of batches, their mechanical behaviour would be similar despite the testing environmental conditions. As a consequence, the number of required experiments for the rigorous material examination could be significantly reduced. Nevertheless, this method does not suit some pairs with the similar values of ΔT , for instance, the batch of -40°C RH 85% with ΔT of +39°C is eventually closer to ΔT of +65.8°C and +63°C rather than +33°C. Similarly, the behaviour of the batch +80°C RH 0%, so ΔT is -24.2°C, is close to +40°C RH 50% with the ΔT -17°C instead of +23°C RH 85%. The authors did not find any clear explanation for such divergence. Miri et al. **[176]** and Arhant **[21]** likewise report the importance of ΔT as a criterion for the characterisation, and demonstrated results are in agreement with this study. Nevertheless, more accurate analysis is required in order to confirm or to reject T as a criterion for rapid mechanical characterisation.





Figure II-53: Shear stress at yield of PA6 GF as a function of environmental conditions and $T_a - T$

Figure II-54: Tendency for change of true shear stress at yield of PA6 GF as a function of $T_g - T$

The yield stress rises along with the increase of ΔT (Figure II-53 – Figure II-54). The elastic domain is, however, hardly pronounced in hot and humid conditions. It can be attributed to the fact that the polymer does not possess a distinct yield point [58] and the divergence of results takes place. Comparable decrease of a linear part of the "stress-strain" slopes as a function of absorbed water by PA6 and testing temperature is also reported by [58], though the moisture impact is assumed to be more severe. The authors show the modifications of the yield stress and the tensile modulus affected by testing temperature (from +4°C to +80°C) and water content (from 0% to 9.3%). And, equivalently, the latter has a stronger effect with the following drop of properties up to 90%. It is associated with more significant mobility of polymer chains. Le Gac et al. [58] also suggest using a Kambour equation [177] to describe the yield shear stress of the composite with the only limitation to the glassy state:

$$\sigma_{12 v true} = A(T_q - T) + B \tag{40}$$

Parameters A and B are the polymer-dependent experimental factors. Despite the divergence of points, the moisture- and temperature-dependent mechanical behaviour seems to follow the trend, demonstrated in Figure II-54. Even though the relation is not found to be validated for the viscous material state, it is included in the Kambour slope (Figure II-54).



Figure II-55: Shear modulus of PA6 GF as a function of environmental conditions and $T_g - T$

The shear modulus is consistent with the in-plane shear yield stress. Figure II-55 and Figure II-56 describe the pattern of modified in-plane shear modulus as a function of ΔT . It is evident that the G_{12} has a tendency to increase with the rising value of ΔT . This signifies that the plasticisation occurs faster in hot and humid conditions as compared to cold and dry environment at the same strain rate. The results are consistent with the studies of [52,78] on a neat PA6, which demonstrate the significant drop of tensile strength and modulus as a result of increasing water content in the material. What is more, the elastic domain tends to diminish faster at RH 85% than in standard humid condition RH 50% or dry RH 0% due to higher ΔT . High temperature increases such effect. Kambour slope (Figure II-56) is applied to illustrate a tendency to a progression of shear modulus over ΔT .



Figure II-56: Change of PA6 GF shear modulus as a function of $T_g - T$

Strain rate effect

Mechanical behaviour of the matrix PA6 is known for its strain-rate dependence at a fixed RH condition. The strain-rate effect can, therefore, modify the temperature within specimens. Thus, the shear strength and strain of the composite are also susceptible to vary. In order to verify such suggestion, several batches are tested with the deformation rate of 0.5 mm/min, four times slower than the velocity used for the whole experimental campaign. Only extreme hot and humid conditions, as RH 85% and +80°C, are selected due to the long test duration. These environmental conditions ensure the viscous material state during the experiments resulting from the T_g exceeding. As a consequence, the possible variations in mechanical behaviour are more likely to be noticed in the chosen conditions.



Figure II-57: Comparison of testing velocity 2 mm/min and 0.5 mm/min for PA6 GF

The global composite material state at the slow testing rate of 0.5 mm/min and its comparison to 2 mm/min is illustrated in Figure II-57. The most noticeable difference occurs in the plastic domain at the high shear strain. Regarding the elastic domain, some distinctions of shear strength and modulus appear, as shown in Table II-11.

Likewise with 2 mm/min, the mechanical properties at slow velocity demonstrate the same tendency to decrease **[178]**. Comparing shear moduli of the standard strain rate to the low, one can notice the slight change of the latter. However, no clear relation between environmental conditions and the strain rate can be established due to the small number of tests and only two loading rates. Graphical representations of shear moduli and yield shear stress as a function of ΔT and loading rate are proposed in Appendix D (Figure VII-19 – Figure VII-20).

Each experimental batch contained a reference specimen in order to monitor the loss or gain of moisture content over testing time. The results are plotted and presented in Appendix D for PA6 GF (Figure VII-19 – Figure VII-20).

Environmental condition	Strain rate $\dot{\varepsilon}$ (s ⁻¹)					
	for 2 mm/min	for 0.5 mm/min				
+80°C/RH 0%	0.189 10-3	0.489 10-4				
+80°C/RH 50%	0.191 10-3	0.503 10-4				
+23°C/RH 85%	0.192 10-3	0.459 10-4				
+40°C/RH 85%	0.199 10-3	0.473 10-4				
+80°C/RH 85%	0.197 10-3	0.506 10-4				

Table II-10: Synthesis of strain rate at studied environmental conditions for PA6 GF

Characterisation of specimen damage

Looking at the ruptured specimens, it is possible to visually estimate the material state – either solid (Figure II-59) or viscous (Figure II-60). The plastic deformation is obvious along with the ductile rupture in the last figure when the simply elastic deformation and the brittle fracture occur in the first case. When ΔT is close to 0°C, a coupled problem is to appear, however, the specimens still tend to the brittle rupture rather than ductile.



Figure II-58: Fibre reorientation of PA6 GF as a function of true shear strain at a) +23°C, b) +40°C and c) +80°C



Figure II-59: Tensile-ruptured specimens PA6 GF of 45° at - 40°C and RH 0%. ΔT is -95.8°C



Figure II-60: Tensile-ruptured specimens PA6 GF of 45° at +80°C and RH 85%. ΔT is +81°C

				45°		0°				90°		
RH	Temp	G ₁₂ MPa	σ _{12 y true} MPa	$\sigma_{12 true max}$ MPa	Y12 true max [%]	E ₁₁ MPa	σ _{11 true max} MPa	ν ₁₂ (-)	E ₂₂ MPa	$\sigma_{22 true max}$ MPa	ν ₂₁ (-)	
0%	-40°C	4769 ±119	6.5 ±0.7	151.2 ±5.9	32.3 ±2.1	_	_	_	_	_	_	
	-10°C	4350 ±163	6.5 ±0.7	130.9 ±8.1	33.3 ±0.4	-	_	-	-	-	-	
	+23°C	3843 ±158	6.3 ±1.3	125.6 ±3.6	41.4 ±5.5	-	-	-	-	-	-	
	+40°C	3527 ±577	6.1 ±3.5	114.4 ±9.3	43.0 ±3.8	-	-	-	-	-	-	
	+80°C	1223 ±109	2.8 ±0.2	116.6 ±7.4	65.3 ±7.1	-	-	_	Ι	-	_	
*	+80°C	1514 ±319	2.0 ±2.8	98.7 ±14.5	54.2 ±8.1	_	_	_	I	-	_	
	-40°C	5071 ±321	6.8 ±1.2	111.3 ±6.9	26.8 ±2.7	20122 ±174	471.2 ±26.4	-	20136 ±1421	410.6 ±177.7	-	
	-10°C	4129 ±111	5.6 ±0.3	104.6 ±2.6	33.5 ±1.4	-	_	-	-	-	-	
50%	+23°C	2383 ±444	3.6 ±1.3	104.0 ±2.8	52.1 ±2.9	21126 ±2570	376.6 ±8.6	0.058 ±0.011	20530 ± 1862	318.6 ±131.7	0.078 ± 0.045	
	+40°C	1750 ±40	3.3 ±0.2	96.8 ±7.1	55.8 ±5.4	-	-	_	-	-	_	
	+80°C	878 ±167	3.4 ±1.7	95.5 ±1.9	66.4 ±2.0	19902 ±182	281.6 ±77.3	0.073 ± 0.050	20262 ±702	285.6 ± 62.9	0.072 ± 0.005	
*	+80°C	812 ±93	5.2 ±0.6	81.6 ±8.6	57.7 ±7.5	19527 ±609	291.3 ± 20.9	0.055 ± 0.007	18973 ±237	266.1 ± 67.8	0.075 ± 0.027	
	-40°C	5055 ±160	7.7 ±3.4	112.7 ±9.0	30.4 ± 3.9	-	-	-	-	-	-	
85%	-10°C	3080 ±991	6.6 ±2.2	93.9 ±2.6	35.5 ±1.8	-	-	_	-	-	_	
	+23°C	1332 ±93	2.9 ±0.3	86.9 ±7.6	54.6 ±8.9	-	-	_	-	-	_	
*	+23°C	1386 ±316	2.8 ±1.3	67.3 ±8.5	40.0 ±8.6	-	-	-	-	-	-	
	+40°C	1253 ±253	2.4 ±0.2	80.3 ±9.2	53.9 ±5.9	-	_	-	-	_	-	
*	+40°C	1135 ±187	3.0 ±2.0	66.0 ±18.9	42.4 ±15.6	-	-	_	_	-	_	
	+80°C	722 ±151	3.9 ±2.8	74.6 ±5.9	58.9 ±4.8	-	-	_	_	-	_	
*	+80°C	621 ±30	4.9 ±0.3	86.7 ±11.5	/3.7 ±20.6	-	-	-	-	-	-	

Table II-11: Mechanical properties of PA6 GF subjected to humidity and temperature. The asterisk * stands for the low strain rate of 0.5 mm/min

Specimens of 45° stacking sequence undergo the fibre reorientation while testing (Figure II-58), particularly of long duration. Such adjustment may cause a change in mechanical parameters if the new fibre orientation is taken into consideration at each time-step of a test. Hence, it is of great interest to delve into this search problem, not treated in this work.

II.4.5. Elastic and ultimate properties of PPS CF

Longitudinal and transverse orientations

Despite the theoretical uniformity of the longitudinal and transverse properties in a balanced woven laminate, the actual mechanical response may slightly differ due to the manufacturing process of the material. Stress – strain curves in Figure II-61 demonstrate such behaviour for PPS CF in the dry state, corresponding to RH 0%, at varying testing temperatures.

With regard to the impact of temperature on mechanical properties, the modulus of elasticity remains broadly unchanged over the entire tested temperature range. However, there is a slight increase at +120°C (Table II-12 at the end of the section). Similar changes are also highlighted in the publications by Vieille et al. **[43,111]**. The comparison of longitudinal and transverse orientations shows that the properties in warp direction (longitudinal) are slightly higher than that of the weft. This difference can be explained by the weaving method: the weft yarns are subjected to undulations in order to create a weaving pattern while the warp yarns remain fixed and in tension. Although stresses and deformations at break seem to decrease above +90°C, the ultimate values cannot be considered and compared since the failure is not permanently occurring in the narrowest cross-section. The experimental results are outlined in Table II-12.

During tensile tests, specimens undergo a brittle fracture that propagates perpendicularly to the loading axis. Some illustrations of ruptured specimens can be found in Appendix D for PPS CF.



Figure II-61: Tensile mechanical behaviour of the composite PPS CF in the dry state

Shear orientation

Characterisation of environmental impact

Tensile tests on 45°-oriented specimens, presented in Figure II-62, show the moisture and temperature impacts on linear and non-linear domains. Around +90°C, +105°C and +120°C the composite is in the ductile state comprising a plastic zone during tests. Consequently, the shear modulus decreases beyond +90°C in a

significant manner. It is also interesting to mention a remarkable increase of total deformation with higher temperature.

Although the water content is moderate within the specimens, we can note a visible impact of moisture on the non-linear part of the stress – strain curves: deformations at failure are generally higher at HR 85% than at HR 0% while stress levels are lower. Thus, as mentioned in the previous sections, the moisture impact can serve for the investigation of elastoplastic behaviour in perspective.

In Figure II-63 and Figure II-64 one can see the evolution of yield shear stress and shear modulus as a function of $T_g - T$. Their increment with the testing temperature decrease is apparent. Vicinity to T_g , which is near +90°C according to [164], seems to change the slope between data points of shear modulus. However, we suggest that a confirmation or rejection of such material property could be realised with a greater number of tests and T_g verification. However, the reduction of shear modulus with higher temperature corresponds to results in [49,109].



Figure II-62: Shear mechanical behaviour of the composite PPS CF in the dry state and at RH 85%



Figure II-63: Shear stress at yield of PPS CF as a function of environmental conditions and $T_g - T$



Figure II-64: Shear modulus of PPS CF as a function of environmental conditions and $T_g - T$

As for the moisture impact, its influence appears to be more important for the yield shear stress than the shear modulus. According to the bibliographic study, immersion in hot water [115] and exposure to RH 75% [26] contribute to the changes of tensile strength. The proposed experimental results appear to be in accordance. Computed mechanical properties for all temperatures and RH are presented in Table II-12.

Specimen damage

The temperature effect becomes enhanced by fibre reorientation while testing, as also observed by Vieille et al. **[43,111]**. The reorientation effect is clearly visible on the specimens in their post-mortem state. The fracture surfaces of specimens are consistent with the fibre reorientation; however, the type of crack is rather brittle for $+23^{\circ}$ C (Figure II-65); on the other hand, it has a ductile nature at $+120^{\circ}$ C (Figure II-66). Some other illustrations can be found in Appendix D in PPS CF section.

		45°				0°			90°		
RH	Temp	G ₁₂ MPa	σ _{12 y true} MPa	σ _{12 true max} MPa	γ12 true max %	E ₁₁ MPa	σ _{11 true max} MPa	ν ₁₂ (-)	E ₂₂ MPa	σ _{22 true max} MPa	ν ₂₁ (-)
	±23°C	3480	15.7	124.7	28.2	54104	648.3	0.036	53443	592.8	0.030
	723 C	±109	±4.8	± 8.7	±3.3	±1386	±33.3	± 0.024	±1089	± 20.7	± 0.005
	±40°C	2536	7.8	105.8	52.7	53703	594.3	0.037	55016	545.9	0.052
00/-	140 C	± 280	±2.3	±12.6	±11.3	± 3033	±1.9	± 0.009	±2215	±36.2	± 0.009
070	±1059C	1222.	6.6	138.2	81.5	53003	513.7	0.046	49704	490.7	0.021
	+105 C	±165	± 2.0	± 30.4	±12.2	± 2277	±27.5	± 0.037	±2455	±7.6	± 0.006
	110000	753	5.6	134.7	88.5	58013	486.6	0.145	55848	457.2	0.119
	±120 C	±160	±4.7	± 47.8	±14.6	± 6672	±10.0	± 0.023	±2234	±12.1	± 0.001
	±23°C	3477	12.1	106.7	25.5						
	125 C	±190	± 5.0	±8.7	±1.8	_	_	_	_	_	-
	±40°C	2506	9.6	116.9	63.1						
850/	140 C	±194	±1.4	±46.3	±30.3	_	_	_	_	_	-
0570	±105°C	1207	4.8	135.2	85.0						
	105 C	±192	±2.2	±22.5	±8.2	_	_	_	_	_	-
	±120%C	564	6.3	153.7	93.7						
	1120 C	± 50	±1.3	±23.1	±4.5	_	_	_	_	_	-

Table II-12: Mechanical properties of PPS CF subjected to humidity and temperature



Figure II-65: Tensile-ruptured specimens of 45° at +23°C and RH 0%



Figure II-66: Tensile-ruptured specimens of 45° at +120°C and $$\rm RH\ 0\%$$

II.4.6. Elastic and ultimate properties of PA6

The understanding of matrix response is necessary in order to thoroughly analyse the mechanical response of the composite PA6 GF. An experimental campaign at the same environmental and testing conditions as for PA6 GF is carried out. As in the case of the composite, obtained results are shown in "true" values due to large strains following the same computation methodology.

Environmental impact

Mechanical behaviour of the neat matrix PA6 is strongly related to the testing environment, as illustrated in Figure II-67 – Figure II-69. We are principally interested in the evolution of elastic properties; hence, the ultimate properties in viscous state are not representative due to the intentional trimmed stress – strain curves at the early stages of the neck formation. In addition, the traction of the majority of specimens is ended at 200% of crosshead displacement due to the limited height of the climatic chamber.



Figure II-67: Mechanical response of PA6 at RH 0%

When the testing temperature is below T_g , the polymer behaves like hard and strong, which is typical for thermoset. Once it is exceeded, the specimens become tough. Along with the final strain hardening, this quality belongs to conventional thermoplastic matrices. Hence, as a result of in-service conditions, the state of the matrix changes affecting physical and mechanical properties. Despite the absence of continuous reinforcing fibres, the results are consistent with the matrix-dominated shear properties of PA6 GF (section II.4.4).


Figure II-68: Mechanical response of PA6 at RH 50%



Figure II-69: Mechanical response of PA6 at RH 85%

In order to analyse the elastic parameters, we compute yield tensile stress and tensile modulus as a function of temperature and RH. As for the yield stress, its estimation is not sufficiently satisfying due to the difficulties for emphasising the elastic part. It is especially challenging when PA6 is in the viscous state because of the almost immediate plastic deformation. This is one of the reasons for an important divergence of experimental points for RH 0% (Figure II-70). The other problem may arise from the frequency of data acquisition during the experimental testing. Thus, it can be possible that the number of points, defining the linear domain, is not sufficient for an accurate evaluation of the yield tensile stress. One more issue may consist in the fact of using the maximum value of the coefficient of determination R^2 instead of a fixed. The demonstrated results serve for a global view of the matrix properties, although, they are not used for the numerical modelling in Chapter III.



Figure II-70: Tensile stress at yield of PA6 as a function of environmental conditions and $T_q - T$



Figure II-71: Elastic modulus of PA6 as a function of environmental conditions and $T_g - T$

As for the elastic modulus, its prediction over ΔT is more accurate. In Figure II-71 – Figure II-72 one can see that the decrease of testing temperature along with changing RH level causes the rise of modulus. Kambour equation is employed, and it demonstrates the coherence with the shear behaviour of the composite as shown in Figure II-56.

The experimental data of elastic modulus and Poisson's ratio at all testing conditions is consequently used as the input parameters for numerical simulations in Chapter III. They are outlined in Table II-13.



Figure II-72: Tendency for change of elastic modulus of PA6 as a function of $T_q - T$

Strain rate effect

As in the case with PA6 GF, the use of slower loading rate at critical environmental conditions does not show the expected impact on stress – strain response (Figure II-73). However, Dau [23] demonstrates a significant change of mechanical properties with relation to crosshead velocity. We suggest that more various loading rates are necessary to be tested in order to reveal modifications in mechanical behaviour. The relation of tensile modulus and yield stress to ΔT and cross-head velocity are graphically presented in Appendix D.

The deviation of the mass of reference specimens is illustrated in Appendix D in Figure VII-41.



Figure II-73: Comparison of testing velocity 2 mm/min and 0.5 mm/min for PA6

$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	RH	Temp	<i>E</i> MPa	σ _{y true} MPa	v (-)	έ s ⁻¹
$\begin{array}{c c c c c c c c c c c c c c c c c c c $			3773	23.7	0.386	
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$		-40°C	± 90	±12.4	± 0.001	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $			3825	13.6	0.384	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $		-10°C	±138	±10.7	±0.01	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $			3934	32.5	0.463	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	0%	+23°C	±220	± 6.5	±0.046	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $		1 4000	3831	21.7	0.455	
$ * \frac{+80^{\circ}C}{+30^{\circ}C} \frac{1177}{\pm 253} \frac{10.3}{\pm 5.7} \frac{0.321}{\pm 0.008} \frac{0.377 10^{-3}}{0.377 10^{-3}} \\ + \frac{+80^{\circ}C}{\pm 126} \frac{\pm 253}{\pm 6.7} \frac{\pm 0.008}{\pm 0.057} \frac{0.827 10^{-4}}{0.827 10^{-4}} \\ \frac{-40^{\circ}C}{\pm 126} \frac{\pm 26.7}{\pm 12.0} \frac{\pm 0.041}{\pm 0.041} \\ \frac{-40^{\circ}C}{\pm 59} \frac{\pm 12.0}{\pm 0.1} \frac{\pm 0.041}{\pm 0.050} \\ \frac{+23^{\circ}C}{\pm 192} \frac{\pm 2.2}{\pm 2.2} \frac{\pm 0.046}{\pm 0.046} \\ \frac{+40^{\circ}C}{\pm 131} \frac{\pm 12.5}{\pm 0.7} \frac{0.537}{\pm 0.018} \\ \frac{+80^{\circ}C}{\pm 105} \frac{\pm 4.0}{\pm 105} \frac{\pm 0.018}{\pm 4.0} \frac{0.391 10^{-3}}{0.391 10^{-3}} \\ \frac{+80^{\circ}C}{\pm 105} \frac{\pm 1.4}{\pm 1.4} \frac{\pm 0.044}{\pm 0.040} \frac{0.391 10^{-3}}{0.885 10^{-4}} \\ \frac{-40^{\circ}C}{\pm 106} \frac{\pm 135}{\pm 3.4} \frac{\pm 0.019}{\pm 0.019} \\ \frac{-40^{\circ}C}{\pm 106} \frac{\pm 135}{\pm 3.1} \frac{5.4}{\pm 0.033} \frac{0.335 10^{-3}}{0.335 10^{-3}} \\ \frac{+23^{\circ}C}{\pm 175} \frac{\pm 1.1}{\pm 3.1} \frac{\pm 0.033}{\pm 0.033} \frac{0.335 10^{-3}}{0.335 10^{-3}} \\ \frac{+23^{\circ}C}{\pm 1135} \frac{5.4}{5.4} \frac{0.493}{0.0314} \frac{0.391 10^{-3}}{0.388 10^{-4}} \\ \frac{+40^{\circ}C}{\pm 16} \frac{\pm 12.6}{\pm 2.0} \frac{\pm 0.014}{\pm 0.014} \frac{0.834 10^{-4}}{0.834 10^{-4}} \\ \frac{+40^{\circ}C}{\pm 16} \frac{\pm 1.6}{\pm 1.3} \frac{\pm 0.025}{\pm 0.017} \frac{0.369 10^{-3}}{0.369 10^{-3}} \\ \frac{+80^{\circ}C}{\pm 16} \frac{526}{\pm 1.9} \frac{10.2}{\pm 0.037} \frac{0.374 10^{-3}}{0.374 10^{-3}} \\ \frac{+80^{\circ}C}{\pm 16} \frac{526}{\pm 1.2} \frac{10.2}{\pm 0.027} \frac{0.966 10^{-4}}{0.966 10^{-4}} \\ \end{array}$		+40°C	±157	±12.0	± 0.032	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $		1 9090	1177	10.3	0.521	0 277 10 3
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $		+80°C	±253	±5.7	± 0.008	0.377 10-5
* +30°C ± 126 ± 6.7 ± 0.057 0.82710^{-1} -40°C $\frac{4234}{\pm 87}$ 17.3 0.353 ± 0.041 10^{-1} -10°C $\frac{3923}{\pm 59}$ 8.4 0.404 ± 0.057 0.82710^{-1} 50% $\pm 23°C$ $\frac{1830}{\pm 192}$ 8.4 0.404 ± 0.050 $\pm 23°C$ ± 192 ± 2.2 ± 0.046 ± 192 ± 2.2 ± 0.046 $\pm 40°C$ ± 131 12.5 0.537 0.39110^{-3} $\pm 80°C$ ± 135 ± 4.0 ± 0.044 0.39110^{-3} $\pm 80°C$ ± 102 ± 2.2 ± 0.046 0.39110^{-3} * $\pm 80°C$ ± 73 ± 1.4 ± 0.044 0.88510^{-4} * $\pm 80°C$ ± 73 ± 1.4 ± 0.044 0.88510^{-4} * $\pm 40°C$ ± 189 ± 3.4 ± 0.019 0.362 * $\pm 23°C$ ± 189 ± 3.4 0.0193 0.33510^{-3}	*	1 8090	1151	9.5	0.486	0.927.10-4
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	4.	+80°C	±126	±6.7	± 0.057	0.827 10 1
$\begin{array}{c c c c c c c c c c c c c c c c c c c $		40°C	4234	17.3	0.353	
$\begin{array}{c c c c c c c c c c c c c c c c c c c $		-40 C	±87	±12.0	±0.041	
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$		-10°C	3923	8.4	0.404	
$\begin{array}{c c c c c c c c c c c c c c c c c c c $		-10 C	±59	±0.1	± 0.050	
120 C ± 192 ± 2.2 ± 0.046 $\pm 40^{\circ}$ C ± 131 12.5 0.537 $\pm 40^{\circ}$ C ± 43 ± 0.7 ± 0.018 $\pm 80^{\circ}$ C ± 105 ± 4.0 ± 0.046 $\pm 80^{\circ}$ C ± 131 ± 0.7 ± 0.018 * $\pm 80^{\circ}$ C ± 105 ± 4.0 ± 0.040 * $\pm 80^{\circ}$ C ± 735 ± 4.0 ± 0.040 * $\pm 80^{\circ}$ C ± 73 ± 1.4 ± 0.044 -40° C ± 73 ± 1.4 ± 0.044 $0.885 10^{-4}$ -40° C ± 73 ± 1.4 ± 0.044 $0.885 10^{-4}$ -40° C ± 73 ± 1.4 ± 0.044 $0.885 10^{-4}$ -40° C ± 189 ± 3.4 ± 0.019 $0.885 10^{-4}$ $\pm 263^{\circ}$ ± 1.2 ± 0.025 $0.335 10^{-3}$ $0.335 10^{-3}$ $*$ $\pm 23^{\circ}$ C ± 75 ± 3.1 ± 0.033 $0.335 10^{-3}$	50%	+23°C	1830	8.6	0.485	
$* \frac{+40^{\circ}C}{+40^{\circ}C} \frac{1131}{\pm 43} \frac{12.5}{\pm 0.7} \frac{0.537}{\pm 0.018}$ $\frac{+40^{\circ}C}{\pm 105} \frac{143}{\pm 10.7} \frac{10.018}{\pm 0.040} \frac{0.391 10^{-3}}{0.391 10^{-3}}$ $\frac{+80^{\circ}C}{\pm 105} \frac{1105}{\pm 4.0} \frac{10.040}{\pm 0.040} \frac{0.391 10^{-3}}{0.391 10^{-3}}$ $\frac{+80^{\circ}C}{\pm 73} \frac{11.4}{\pm 1.4} \frac{10.044}{\pm 0.044} \frac{0.885 10^{-4}}{0.885 10^{-4}}$ $\frac{-40^{\circ}C}{\pm 189} \frac{13.4}{\pm 3.4} \frac{10.019}{\pm 0.019}$ $\frac{-40^{\circ}C}{\pm 106} \frac{11.2}{\pm 1.2} \frac{10.025}{\pm 0.025}$ $\frac{+23^{\circ}C}{\pm 75} \frac{13.1}{\pm 3.1} \frac{10.033}{\pm 0.033} \frac{0.335 10^{-3}}{0.335 10^{-3}}$ $\frac{+23^{\circ}C}{\pm 26} \frac{871}{\pm 2.0} \frac{11.3}{\pm 0.014} \frac{0.834 10^{-4}}{0.834 10^{-4}}$ $\frac{+40^{\circ}C}{\pm 16} \frac{764}{\pm 1.3} \frac{9.1}{\pm 0.025} \frac{0.369 10^{-3}}{0.369 10^{-3}}$ $\frac{+40^{\circ}C}{\pm 16} \frac{781}{\pm 9.1} \frac{9.1}{0.525} \frac{0.330 10^{-4}}{0.331 10^{-3}}$ $\frac{+80^{\circ}C}{\pm 16} \frac{516}{\pm 1.2} \frac{10.2}{\pm 0.037} \frac{0.374 10^{-3}}{0.374 10^{-3}}$	30%		±192	±2.2	±0.046	
* ± 43 ± 0.7 ± 0.018 * $\pm 80^{\circ}$ C 745 9.9 0.531 $0.391 10^{-3}$ * $\pm 80^{\circ}$ C ± 105 ± 4.0 ± 0.040 $0.391 10^{-3}$ * $\pm 80^{\circ}$ C ± 105 ± 4.0 ± 0.040 $0.391 10^{-3}$ * $\pm 80^{\circ}$ C ± 73 ± 1.4 ± 0.044 $0.885 10^{-4}$ * $\pm 40^{\circ}$ C ± 73 ± 1.4 ± 0.044 $0.885 10^{-4}$ * -40° C ± 189 ± 3.4 ± 0.019 $0.885 10^{-4}$ * -10° C 2639 5.7 0.388 0.019 * -10° C 2639 5.7 0.388 $0.335 10^{-3}$ * -10° C ± 106 ± 1.2 ± 0.025 $0.335 10^{-3}$ * $\pm 23^{\circ}$ C ± 75 ± 3.1 ± 0.033 $0.335 10^{-3}$ * $\pm 23^{\circ}$ C ± 75 ± 3.1 ± 0.033 $0.369 10^{-3}$		+40°C	1131	12.5	0.537	
$\begin{array}{c c c c c c c c c c c c c c c c c c c $			±43	±0.7	±0.018	
$\begin{array}{c c c c c c c c c c c c c c c c c c c $		+80°C	745	9.9	0.531	0.391 10-3
* +80°C 808 ±73 3.7 ±1.4 0.537 ±0.044 0.885 10.4 -40°C 4940 ±189 1.9 0.362 ±0.019 0.885 10.4 85% -10°C 2639 ±106 5.7 0.388 ±0.025 +23°C 1135 5.4 0.493 0.335 10.3 * +23°C 871 11.3 0.511 0.884 10.4 +40°C 764 10.5 0.518 0.369 10.3 * +40°C 781 9.1 0.525 0.830 10.4 * +80°C 516 12.4 0.505 0.374 10.3 * +80°C 526 10.2 0.496 0.966 10.4			±105	±4.0	±0.040	
$\begin{array}{c c c c c c c c c c c c c c c c c c c $	*	+80°C	808	3.7	0.537	0.885 10-4
-40°C $\frac{4940}{\pm 189}$ $\frac{11.9}{\pm 3.4}$ $\frac{0.362}{\pm 0.019}$ 85% -10°C $\frac{2639}{\pm 106}$ 5.7 0.388 +23°C $\frac{1135}{\pm 75}$ 5.4 0.493 $0.335 10^{-3}$ * +23°C $\frac{871}{\pm 75}$ ± 3.1 ± 0.033 $0.335 10^{-3}$ * +23°C $\frac{871}{\pm 26}$ ± 2.0 ± 0.014 $0.834 10^{-4}$ +40°C $\frac{764}{\pm 16}$ 10.5 0.518 $0.369 10^{-3}$ * +40°C $\frac{781}{\pm 25}$ 9.1 0.525 $0.830 10^{-4}$ +80°C 516 12.4 0.505 $0.374 10^{-3}$ * +80°C $\frac{526}{\pm 16}$ 10.2 0.496 $0.966 10^{-4}$			±73	±1.4	±0.044	
85% ± 189 ± 3.4 ± 0.019 85% -10° C 2639 5.7 0.388 ± 106 ± 1.2 ± 0.025 $\pm 23^{\circ}$ C 1135 5.4 0.493 $0.335 10^{-3}$ * $\pm 23^{\circ}$ C 871 11.3 0.511 $0.834 10^{-4}$ * $\pm 23^{\circ}$ C 871 11.3 0.511 $0.834 10^{-4}$ * $\pm 23^{\circ}$ C 871 11.3 0.511 $0.834 10^{-4}$ * $\pm 40^{\circ}$ C 764 10.5 0.518 $0.369 10^{-3}$ * $\pm 40^{\circ}$ C 781 9.1 0.525 $0.830 10^{-4}$ * $\pm 40^{\circ}$ C 516 12.4 0.505 $0.374 10^{-3}$ * $\pm 80^{\circ}$ C 516 12.4 0.505 $0.374 10^{-3}$ * $\pm 80^{\circ}$ C 526 10.2 0.496 $0.966 10^{-4}$		-40°C	4940	11.9	0.362	
85% -10°C $2639 \\ \pm 106 \\ \pm 106 \\ \pm 1.2 \\ \pm 0.025$ +23°C 1135 \\ \pm 75 \\ \pm 75 \\ \pm 3.1 \\ \pm 0.033 \\ \pm 26 \\ \pm 26 \\ \pm 2.0 \\ \pm 2.0 \\ \pm 0.014 \\ \pm 0.014 \\ 0.834 10^4 \\ 0.830 10^4 \\ 0.830 10			±189	±3.4	±0.019	
$* \frac{\pm 106}{\pm 1.2} \frac{\pm 0.025}{\pm 0.025}$ $+23^{\circ}C \frac{1135}{\pm 75} \frac{5.4}{\pm 3.1} \frac{0.493}{\pm 0.033} \frac{0.335 10^{-3}}{0.335 10^{-3}}$ $* \frac{+23^{\circ}C}{\pm 26} \frac{871}{\pm 2.0} \frac{11.3}{\pm 0.014} \frac{0.834 10^{-4}}{0.834 10^{-4}}$ $+40^{\circ}C \frac{764}{\pm 1.6} \frac{10.5}{\pm 1.3} \frac{0.518}{\pm 0.025} \frac{0.369 10^{-3}}{0.369 10^{-3}}$ $* \frac{+40^{\circ}C}{\pm 25} \frac{781}{\pm 0.9} \frac{9.1}{\pm 0.017} \frac{0.830 10^{-4}}{0.830 10^{-4}}$ $+80^{\circ}C \frac{516}{\pm 1.6} \frac{12.4}{\pm 1.9} \frac{0.505}{\pm 0.037} \frac{0.374 10^{-3}}{0.374 10^{-3}}$ $* \frac{+80^{\circ}C}{\pm 16} \frac{526}{\pm 0.9} \frac{10.2}{\pm 0.037} \frac{0.496}{0.966 10^{-4}}$	85%	-10°C	2639	5.7	0.388	
$* \frac{+23^{\circ}C}{\pm 75} \frac{1135}{\pm 3.1} \frac{5.4}{\pm 0.033} \frac{0.493}{\pm 0.033} = 0.335 10^{-3}$ $* \frac{+23^{\circ}C}{\pm 26} \frac{871}{\pm 26} \frac{11.3}{\pm 20} \frac{0.511}{\pm 0.014} = 0.834 10^{-4}$ $+40^{\circ}C \frac{764}{\pm 16} \frac{10.5}{\pm 1.3} \frac{0.518}{\pm 0.025} = 0.369 10^{-3}$ $* \frac{+40^{\circ}C}{\pm 25} \frac{781}{\pm 0.9} \frac{9.1}{\pm 0.017} = 0.830 10^{-4}$ $+80^{\circ}C \frac{516}{\pm 16} \frac{12.4}{\pm 1.9} \frac{0.505}{\pm 0.037} = 0.374 10^{-3}$ $* \frac{+80^{\circ}C}{\pm 16} \frac{526}{\pm 10.2} = 0.496 = 0.966 10^{-4}$			±106	±1.2	±0.025	
± 75 ± 3.1 ± 0.033 * $\pm 23^{\circ}$ C 871 11.3 0.511 $0.834 \ 10^4$ ± 26 ± 2.0 ± 0.014 $0.834 \ 10^4$ $\pm 40^{\circ}$ C 764 10.5 0.518 $0.369 \ 10^{-3}$ * $\pm 40^{\circ}$ C 764 10.5 0.518 $0.369 \ 10^{-3}$ * $\pm 40^{\circ}$ C 781 9.1 0.525 $0.830 \ 10^{-4}$ * $\pm 40^{\circ}$ C 516 12.4 0.505 $0.374 \ 10^{-3}$ * $\pm 80^{\circ}$ C 516 12.4 0.505 $0.374 \ 10^{-3}$ * $\pm 80^{\circ}$ C 526 10.2 0.496 $0.966 \ 10^{-4}$		+23°C	1135	5.4	0.493	0.335 10-3
* +23°C $\frac{871}{\pm 26}$ 11.3 0.511 0.834 10.4 +40°C $\frac{764}{\pm 16}$ 10.5 0.518 0.369 10.3 * +40°C 781 9.1 0.525 0.830 10.4 +40°C 781 9.1 0.525 0.830 10.4 +40°C 516 12.4 0.505 0.374 10.3 * +80°C 526 10.2 0.496 0.966 10.4			±75	±3.1	±0.033	
$* \frac{\pm 26}{\pm 40^{\circ}\text{C}} \frac{\pm 2.0}{\pm 16} \frac{\pm 0.014}{\pm 0.014} = \frac{\pm 0.014}{\pm 0.014}$ $* \frac{+40^{\circ}\text{C}}{\pm 16} \frac{764}{\pm 1.3} \frac{\pm 0.025}{\pm 0.025} = \frac{0.369 \ 10^{-3}}{\pm 0.025}$ $* \frac{+40^{\circ}\text{C}}{\pm 25} \frac{781}{\pm 0.9} \frac{9.1}{\pm 0.017} = \frac{0.830 \ 10^{-4}}{\pm 0.017} = 1000000000000000000000000000000000000$	*	+23°C	8/1	11.3	0.511	0.834 10-4
$* +40^{\circ}C +40^{\circ}C +16 +10.5 +10.5 +10.518 +10.025 = 0.369 +10^{-3} +16 +10.5 +10.025 = 0.369 +10^{-3} +10^{\circ}C +16 +10.9 +10.017 = 0.830 +10^{-3} +10.017 +10.017 = 0.830 +10^{-3} +10.017 +10.017 = 0.374 +10^{-3} +10.017 = 0.374 +10^{-3} +10.017 = 0.374 +10^{-3} +10.017 = 0.$			±26	±2.0	±0.014	
$* +40^{\circ}C +80^{\circ}C +80^{\circ}C +80^{\circ}C +62 +62 +62 +62 +62 +62 +62 +62 +62 +62$		+40°C	/64 +16	10.5	0.518	0.369 10-3
* +40°C $\frac{781}{\pm 25}$ $\frac{9.1}{\pm 0.9}$ $\frac{0.325}{\pm 0.017}$ $\frac{0.830 10^4}{20.017}$ +80°C $\frac{516}{\pm 16}$ $\frac{12.4}{\pm 1.9}$ $\frac{0.505}{\pm 0.037}$ $\frac{0.374 10^{-3}}{20.0374 10^{-3}}$ * +80°C $\frac{526}{\pm 62}$ $\frac{10.2}{\pm 1.4}$ $\frac{0.496}{200}$ $\frac{0.966 10^{-4}}{200}$			<u> </u>	<u> </u>	±0.025	
* +80°C 516 12.4 0.505 0.374 10 ⁻³ * +80°C 526 10.2 0.496 0.966 10 ⁻⁴	*	+40°C	/81 +25	9.1	0.525 +0.017	0.830 10-4
+80°C 516 12.4 0.303 $0.374 \ 10^{-3}$ * +80°C 526 10.2 0.496 $0.966 \ 10^{-4}$			<u> </u>	12.4	0.505	
* +80°C 526 10.2 0.496 0.966 10.4		+80°C	+16	12.4 +1.0	+0.037	0.374 10-3
* +80°C 520 10.2 0.420 0.966 10.4			526	10.2	0.037	
± 13 ± 10030	*	+80°C	+62	+1 3	+0.030	0.966 10-4

Table II-13: Mechanical properties of PA6 subjected to humidity and temperature. The asterisk * stands for the low strain rate 0.5 mm/min

II.4.7. Conclusions

Carried tensile testing campaigns enable us to reveal the impact of different environmental parameters on the in-plane properties of two composite materials and the matrix, and to analyse material durability. They are related to both the testing environment, represented by temperature and moisture content in materials, and the fibre

orientations in order to assess the fibre and matrix-dominated properties of the composites that vary due to environmental conditions. Hence, all specimens are conditioned prior to experiments.

A great number of experiments on PA6 GF demonstrates a coupled effect of moisture and temperature on the whole range of matrix-dominated mechanical properties. Environmental impact leads to the sudden drop of shear strength, yield shear stress and shear modulus once T_g is exceeded. Rise of temperature and RH induces bigger shear strains, which emphasises the viscous nature of the composite. Some mechanical and physical properties are also reported for longitudinal and transversal fibre orientations. However, no significant thermal and humid impact is found.

As for the neat matrix PA6, its tensile behaviour correlates with matrix-dominated PA6 GF. Rigid and brittle nature of specimens changes for ductile and viscous above T_g . Experiments at high temperatures validate the appearance of plastic strain possessing a strong tendency to increase with greater moisture content. Tensile stresses and modulus demonstrate a comparable to PA6 GF reducing trend towards hot and wet conditions.

The water content does not show any significant effects on PPS CF, although it can be seen that the composite material tends to be more deformable. The in-service temperatures are, however, to be taken into consideration due to significantly affected in-plane shear mechanical properties. Regarding the humid conditions, it is not necessarily recommended to apply the conditioning protocol for the definition of elastic properties; however, it is suggested to condition the composite prior to evaluation of non-linear behaviour. As for the warp and weft properties of the composite material, a slight change in modulus can be observed at +120°C; otherwise in all other cases, the temperature has no significant impact on the longitudinal and the transversal mechanical behaviour. Nevertheless, the validation of the environmental impact on the mechanical behaviour of carbon-fibre reinforced PPS composite allows the construction of a reliable experimental base. Finally, both experimental campaigns also provide a considerable amount of extra information concerning the environmental sensitivity of the shear damage as well as the shear irreversible strains. As mentioned before, it was decided not to present this processed data due to its irrelevance for the bolted joints. However, we provide a summary of the results in Appendix D. This work can contribute to a complete understanding of the behaviour of the composite and to serve for the development of adapted constitutive behaviour laws.

In relation to future work, the demonstrated experimental campaign may be expanded by the application of different strain rates for the matrix and the shear loading for both composites. It is specifically interesting for the PPS CF since no information is found regarding the response of this material under several loading velocities. Such data would serve for more detailed analysis of three given materials since a boundary between quasi-static and dynamic states remains questionable at present. Besides, data acquisition and processing methods can play an important role. For instance, the use of strain gauges, extensometers or high-resolution images may eventually have a slight impact on final results; the type of acquisition method should be considered when high precision is demanded. As for the evaluation of elastic properties, a great number of data in the linear zone may contribute in a more accurate and rigorous evaluation of moduli and yield stresses.

The obtained database on the composite materials and the matrix PA6, which may be enriched in perspective by the data of the neat matrix PPS, is then used for a numerical study of double-scale Representative Volume Elements of the composites as a function of proposed environmental conditions (Chapter III). The particular importance of numerical modelling lies in the computation of out-of-plane characteristics that are complex to obtain experimentally. Consequently, provided experimental and numerical mechanical properties as a function of environmental conditions are necessary for the estimation of mechanical behaviour over time of pre-stressed bolted joints (Chapter IV).

CHAPTER III IDENTIFICATION OF OUT-OF-PLANE ELASTIC PROPERTIES VIA HOMOGENISATION METHOD

Difficult experimental assessment of out-of-plane behaviour and its importance for the design of pre-stressed bolted joints requires another evaluation approach. Numerical computation of out-of-plane properties, based on the elementary constituents of composite material, is thus proposed in the present Chapter. Formerly evaluated matrix properties allow us to estimate the mechanical performance according to suggested environmental conditions. Computation is based on the double-scale homogenisation with the use of two representative volume elements. In order to reproduce a Representative Volume Element of the material, the composite geometry, tow and matrix behaviour are necessary to be analysed at first. Eventually, a whole range of numerical in-plane and out-of-plane mechanical properties enables the validation procedure. It is proposed in order to compare the experimentally obtained in-plane mechanical properties to numerical in-plane ones.

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III.1. BRIEF REVIEW OF EXPERIMENTAL METHODS FOR OUT-OF-PLANE PROPERTIES

The majority of out-of-plane experiments is based on shear loading. Besides, non-destructive testing is also used for the characterisation. The potential methods are the short three-point bending, losipescu test, v-notched rail shear, double notch compression, inclined double shear, torsion and ultrasonic characterisation. Advantages and drawbacks of these tests are presented in the following section.

Short 3 point bending

The present test is based on ASTM D2344 [179] or ISO 14130 [180] and is frequently employed for quality control in the industry. However, only the qualitative measurement of the interlaminar shear strength is eventually provided [181,182]. It is caused by a complex stress state in specimens due to the combination of interlaminar shear stress, contact and high flexural stresses on the upper and lower surfaces. Consequently, invalid failure modes can be observed. Although this does not result in the accurate strength estimation [174,182,183], Schneider et al. [184] achieved an excellent reproducibility of the shear moduli G_{12} , G_{13} , G_{23} during shear test on an interlock composite material with the use of DIC and the short three-point bending. Authors demonstrate that the use of a complete RVE is not always possible due to probable incompatibility with the theory of elasticity.



Figure III-1: Short 3 point bending test in 1-3 plane [185]

losipescu test

The experiment is based on ASTM D5379 **[186]** and also named as a "V-notched beam method". ASTM D5379 defines proper fibre orientations for each shear modulus and describes satisfactory or not failure modes. Although the test is considered as relatively reliable for shear strength estimation **[174]**, other authors demonstrate non-uniform stress distribution in the gauge section **[182,187]**.



Zhou et al. [174] suggest using dummy tabs made of woven fabric prepregs in order to fit a specimen in the test device (Figure III-2). They also prevent a specimen from edge-crushing. However, the evaluation of

interlaminar shear strength involves the use of thick specimens; otherwise, undesirable twisting may occur. In addition, the fixture restricts the geometry of coupons. Thus, the composites with an important RVE are difficult to test.

V-notched rail shear test

The present testing method represents the combination of Iosipescu and two-rail shear tests (Figure III-4). Hence, it reassembles the advantages and the weaknesses of both. The presence of V-notches contributes to a relatively uniform stress state between them, where the gauges are adhered. The problem of edge-crushing is eliminated as well. In addition, high shear stress levels can be achieved **[188,189]**. However, stress concentrations occur where the rails are clamped and the specimen geometry must be large, which is often not convenient for the identification of out-of-plane properties **[188]**. Finally, the test fixture is more appropriate for the definition of in-plane shear properties **[181]**.



Figure III-4: The two- (a) and three-rail (b) shear methods. The specimen (darker grey) is bolted in the fixture and the load is applied to the fixture. For the configuration on the right, laminate is clamped on the opposite edges with a tensile or compressive load applied to the third pair of rails in the centre [181]

Double notch compression and inclined double notch shear

The double notch compression test is regulated by ASTM D3846 **[190]**. Main disadvantage consists in a non-homogeneous stress field over a specimen and severe stress concentrations in the vicinity of notches (Figure III-5) **[181]**. According to **[182]**, the interlaminar shear strength is underestimated for unidirectional composites.



The stress state is rather uniform for the inclined double notch shear test (Figure III-6). It is reached with two superimposed point loads and, thus, the elimination of stress concentrations near notches. However, the shear strength is different from the one measured with Iosipescu test **[181,182]**.

Torsion

The out-of-plane shear can be estimated with the torsional loading of coupons (Figure III-7). Although stress state is non-homogeneous, the pure torsion enables to obtain zero values in the neutral fibre **[184]**. A large geometry of the coupon is advantageous for materials with a big RVE.

By carrying out tests on selected fibre orientations and measuring the angle of twist and transverse shear, Tsai et al. **[191]** determined all shear moduli of a composite. In order to avoid edge effects, that may impact the transverse shear and angle of twist, a specimen should be long enough by reason of strain measurements on the surface and the edge. Schneider et al. **[184]** successfully evaluated out-of-plane shear moduli of a woven composite by proposing a methodology, based on the torsional loading.



Figure III-7: Torsion of a bar [184]

Ultrasonic characterisation method

This approach is described and applied by Dupin [192], Hamonou [1] and Baste [193]. Importance of ultrasonic characterisation is the preservation of mechanical integrity of specimens due to the absence of deterioration. This method provides a link between the propagation velocity of ultrasonic waves within a material to its elastic properties allowing computation of the complete elasticity tensor as mentioned by to Dupin [192].

Hamonou [1] has adopted ultrasonic characterisation in order to experimentally analyse the out-of-plane elastic modulus E_{33} of metals, matrices and orthotropic composite materials. Hamonou demonstrates that the obtained results are satisfactory for isotropic materials with a maximum deviation of 13% from theoretical values, whereas the orthotropic composites demonstrate 40% of the difference. The main difficulty lies in the size of the sensor, which, theoretically, should be of the dimensions of the RVE (to recall, the size of the RVE of PA6 GF is 16×16 mm²). Hence, more sophisticated protocols are required for more accurate results.

Conclusions

The demonstrated experimental methods appear to be the most appropriate for evaluation of the out-ofplane shear moduli G_{13} , G_{23} [182,185]. However, one of the principal interests of the project lies in the estimation of the out-of-plane elastic modulus E_{33} as it is of crucial importance for the preloaded bolted structures (Chapter I section II.2). Due to the complexity of experiments, restrictions to specimen geometry, non-homogeneous stress state, large RVE and minimal thickness of studied composites, the mechanical experimental evaluation of out-of-plane elastic modulus seems to be complex. Furthermore, it is likewise difficult to compute it with the ultrasonic characterisation due to considerable errors, caused by insufficient sensor size. As an alternative solution for determination of E_{33} , it is proposed to identify material properties via homogenisation method by using RVE reconstruction (section III.2) and Finite Element Method (FEM) (section III.3).

III.2. RECONSTRUCTION OF REPRESENTATIVE VOLUME ELEMENT

In this section, we propose a step-by-step reconstruction of representative volume elements of studied composite materials with woven reinforcement. The geometry of tows and fibre contents are analysed with Scanning Electron Microscope, software ImageJ and presented in TexGen.

III.2.1. Structure and image analysis

Microscopic study

To perform numerical simulations on a true RVE, the determination of geometrical parameters within a composite is essential. For an accurate evaluation, a microscopic study is carried out on a Scanning Electron Microscope (SEM) JEOL-JSM-6060LA with a useful dimension of $80 \times 40 \times 50$ mm³ and magnification from 8 to 300000 (Figure III-8). The microscope also has an energy-dispersive X-ray spectroscopy detector for chemical characterisation of specimens. Composite coupons of 20×20 mm² are cut from laminates along longitudinal fibre orientation for subsequent desorption prior to microscopic analysis. Hence, polished cross-sections of weft and warp directions provide all the information required for RVE reconstruction. Both composites, PA6 GF and PPS CF, are analysed. The latter coupons are coated with a thin layer of gold in a sputter-coating to ensure the contrast between the matrix and the fibres.



Figure III-8: Scanning Electron Microscope

For further RVE modelling, the microscopic study is focused on the horizontal and vertical distances between tows, their length and width, fibre area fractions, fibre diameters. To retrieve this information, the different magnifications are set. There are, however, few precautions to be taken:

- The same distance between a specimen and the sensor should be maintained to ensure the reproducibility and avoid the sensor damage;
- The magnification zoom should be the same for all the captured images; the scale may vary upon a requirement;
- The captured images should contain the label information, for instance, magnification, specimen number and name, scale and volt;
- For further image analysis, one shall ensure sharply defined fibres on images when possible.

Some original images are presented in Figure III-9 and Figure III-10 for PA6 GF and PPS CF accordingly. With higher magnifications, we are able to see the fibre distribution within a tow, its diameter and structure integrity, whereas the lower magnifications provides the geometrical shape and dimensions of tows as well as their distributions within a material thickness. All the taken images are consequently analysed as detailed in the next section.



Figure III-9: Microscopic structure of PA6 GF: a) ×900 and b) ×30 magnifications



Figure III-10: Microscopic structure of PPS CF: a) ×700 and b) ×60 magnifications

Image analysis

The geometrical properties are obtained by calibrating the pixels to real distance, indicated on each image. The images are analysed with software ImageJ and Matlab. The latter is used to automate the computations of fibre content in tows of PPS CF. Known magnification and distances in pixels and millimetres allow us to measure the thickness and width of tows. As for the computation of fibre area fraction at the mesoscopic level, the next steps are followed in ImageJ:

- Set a scale of the image;
- Isolate a tow prior to further actions (Figure III-11);
- Convert the RGB image to 8-bit grayscale image (Figure III-12);
- Reduce the noise by applying a filter, for instance, median Gauss filter;
- Apply threshold to images to bifurcate the fibres and the matrix;
- Remove the excessive information;
- Apply watershed function to perform fibre segmentation (Figure III-13);
- Calculate the ratio of black pixels to the total number of pixels in tow, which give the fibre fraction (Figure III-14 – Figure III-15).

Figure III-11: Isolated tow of PA6 GF



There are, however, some assumptions applied:

- Fibres, significantly separated from the main part, are included in the computation of fibre fraction and total tow surface, but do not form an integral tow structure;
- Tow surface is limited to the external fibres, thus not smooth;
- Indents on borders are excluded from the tow surface.

As one can notice from Figure III-11 – Figure III-15, the low magnification provides the information about tow geometrical parameters, but the fibre content appears to be influenced by the extremely tiny distance between fibres. This may result in the artificially elevated fibre fraction. In order to propose more reasonable results, the fibre content is computed for pictures, randomly taken inside tows (at higher magnitude). However, Ruijter **[194]** and Koissin et al. **[195]** indicate the difference of fibre content near the tow side and in the centre. Hence, we propose to consider an average of two magnifications. The same steps, described for computations at tow level, are followed and illustrated in Figure III-16.

The geometrical properties, retrieved from the microscopic analysis, are summarised in Table III-1. The table also gives the average values of fibre content within a tow: 72.44% (average of fibre contents at both magnifications 27 and 900) for PA6 GF and 61.97% for PPS CF. The number of treated images of tows is 50 for PA6 GF at two scales and 35 for PPS CF at one scale. Obtained results are then used for the full RVE reconstruction of both composites, presented in the following section.

Specification	PA6 GF	(ImageJ)	PPS CF (Matlab)		
specification	Scale ×27	Scale ×900	Scale ×700		
Average fibre fraction	71.6 %	73.27 %	61.97 %		
Major axis of tow	2914.	48 μm	2103.92 μm		
Minor axis of tow	202.3	31 µm	139.06 µm		
Horizontal distance between tows	316.2	25 μm	276.42 μm		
Vertical distance between tows	104.9	91 μm	11.74 µm		

Table III-1: Average geometrical properties of RVE PA6 GF and PPS CF



Figure III-16 : Evaluation of fibre content at microscopic scale: a) a proper scale is set for an image; b) unnecessary boundaries are removed; c) conversion to 8-bit grayscale image; d) noise reduction and removed excessive information; e) fibre segmentation; f) calculation of fibre area

III.2.2. Generation of composite geometries

RVE models of woven composite materials are reconstructed using software TexGen according to the local observations with SEM and parameters that are given in Table III-1. TexGen has various modules for textile modelling allowing parameterisation and simple modification of the textile structure. The main challenge during this modelling is to avoid overlapping of the tows as the vertical distance between two consecutive tows is very small. The same model can be constructed with CATIA, but the problem of overlapping of tows remains, whereas in TexGen the overlapping can be seen very clearly and interpolation can be done to the specific control point to remove the surface interference. This is particularly important for the cross-section, employed in this work. Despite the locally observed deformed cross-sections, for instance, Figure III-17, their geometry is not easily describable with parameters, proposed in TexGen [194]. For this reason, an elliptical cross-section is thus used to model the tows. Its advantage consists in the smooth edges that also compensate for small separated

clusters of fibres. The tow path is represented by a b-spline with four controlled points (Figure III-18); however, their number can be increased for better representation of undulation.

Material	Tow cross-section	Thickness	Width	Final fibre content*	Fibre content from data sheets
PA6 GF	Ellipse	0.176 mm	2.914 mm	47.27 %	47 %
PPS CF	Ellipse	0.150 mm	2.104 mm	42.8 %	45 %

Table III-2: RVE properties. *Final fibre content is the multiplication of fibre content in tow by a total volume of tows within the RVE



(a) Global flattening (b) Local flattening



(c) Yarn twisting

Figure III-17: Tow cross-section deformation mechanisms, shown in local (yarn cross-section) coordinate system [194]



Figure III-18: Tow reconstruction in TexGen with tow path and control points

As a real RVE, representing the macroscopic behaviour of the material, the proposed ones comprise the tows linked together by the matrix (light grey colour in Figure III-19). Tow's dimensions are slightly modified in order to adapt the total real fibre content, consisting of tow fibre content and tow volume within the RVE, to the fibre content proposed by supplier **[163,164]**.

Obtained model of woven textile pattern provides Python output, which can be used for computations by the finite element approach. The RVE of PA6 GF is consequently adopted for computations in Cast3M-CEA with the use of finite elements. As for the composite material PPS CF, it is not used for the following study for the reason that the neat specimens of PPS are not available. Hence, the mechanical characteristics of this matrix are unknown for studied environmental conditions, making it impossible to characterise the properties of the tows and the composite.



Figure III-19: RVE reconstruction in TexGen: a) PA6 GF; b) PPS CF

III.2.3. Conclusions

The particular interest of this section is to evaluate the real structure of composites. The microscopic study with SEM enables the evaluation of tow true dimensions and fibre contents. What is more, one highlights that the fibre content near tow edges is lower than in the centre. Hence, for rigorous research, the necessity of microscopic analysis at different magnifications is evident.

Geometries of tows and their positions through the thickness are likewise inspected. The non-uniform crosssection makes it difficult to model each tow independently. Therefore, an elliptical cross-section is employed. Two RVE models are generated in conformity with the tow spacing, geometry and total fibre content. Nevertheless, for the further numerical analysis, presented in section III.3, the only RVE of PA6 GF is used. The numerical mechanical properties of PPS CF cannot be analysed due to the unavailability of its matrix properties at the studied environmental conditions, described in Chapter II.

III.3. DOUBLE-SCALE HOMOGENISATION

Out-of-plane mechanical properties are of a crucial significance for bolted structures. Similarly to in-plane experimental results, they may be prone to diminish with the rise of in-service temperature and humidity. By reason of small thickness of composites and, thus, experimental difficulties, we suggest adopting a numerical approach with the use of a double-scale homogenisation in order to evaluate the whole range of elastic mechanical properties of the composite using FEM via Cast3M-CEA. Known matrix properties under varying weather conditions allow identification of the composite behaviour for the identical environment. The validation of numerical properties is achieved by correlating to experimental in-plane ones.

III.3.1. Concept of homogenisation

As one can perceive from the image analysis in the previous section, the behaviour of composite materials may be defined at different scales: macro, meso and micro (Figure III-20). Ha **[16]** and Mbacke **[17]** propose the next clarification for each scale:

- Macro level identifies the 3D composite geometry. One can study the global material deformations and occurring stresses at this scale;
- Meso-level corresponds to the scale of tows. It defines the internal structure of tows, fibre volume fraction inside tows, fibre orientation;
- Micro level corresponds to the scale of fibres and defines fibre arrangement inside the tow RVE.
 Fibre integrity, its diameter and interactions between fibres can be studied.



Figure III-20: Different scales [17]

Figure III-20 demonstrates that the composite material can be seen as a homogeneous RVE. This is, however, valid for structural components, analysed at the macro scale. The study at this level does not consider the impact of fibre and tow dimensions, their positions and interactions, presence of micro-damages, impurities, etc. Although the macro-level analysis is known for its relative simplicity, the macroscopic properties can be strongly impacted by the lack of information, for instance, properties of constituents that are non-identifiable at the macro level. Nevertheless, the heterogeneities can be taken into consideration at finer scales: meso and micro. The challenge would consist in the random fibre distribution within tows, imperfect tows positioning through the composite, inconsistent fibre and tow geometry; thus, it is barely possible to consider all the heterogeneities for subsequent structural applications. In order to overpass these difficulties, the homogeneous one, thus, preserving the influence of constituents at the micro-scale. Commonly, the homogenisation is applied to the RVE (Figure III-19) – a minimum volume characterising the material behaviour.

For rigorous evaluation of composite properties under environmental conditions, we propose to apply a double-scale homogenisation method using FEM via Cast3M-CEA. The first homogenisation is performed on the micro-scale (fibres and matrix) corresponding to the tow, while the second one is on the macro-scale (tows and matrix) corresponding to the RVE.

The theoretical background of homogenisation is based on constitutive relations for linear elasticity. From the generalised Hooke's law, the constitutive law for the heterogeneous linear elastic structure is following **[196]**:

$$\overline{\overline{\sigma}} = \mathbb{C}: \overline{\overline{\epsilon}}$$
⁽⁴¹⁾

$$\overline{\overline{\varepsilon}} = \mathbb{S} : \overline{\overline{\sigma}} ; \ \mathbb{S} \coloneqq \mathbb{C}^{-1}$$
(42)

where $\overline{\epsilon}$ and $\overline{\sigma}$ are local strain and stress tensors, \mathbb{C} and \mathbb{S} stand for stiffness and compliance tensors respectively. In order to obtain equivalent structure behaviour from the local (in other words, the homogeneous one from heterogeneous) in an RVE, the next expressions are used [197]:

$$\overline{\Sigma} = <\overline{\sigma} > = \frac{1}{|V|} \int_{V} \overline{\sigma} dV \tag{43}$$

$$\overline{\overline{E}} = <\overline{\overline{\epsilon}} > = \frac{1}{|V|} \int_{V} \overline{\overline{\epsilon}} dV$$
(44)

where $\langle \overline{\sigma} \rangle$ and $\langle \overline{\epsilon} \rangle$ correspond respectively to average microscopic stress and strain throughout the volume V that is the actual RVE. $\overline{\Sigma}$ and \overline{E} are the macroscopic stress and strain.

Considering the mechanical impact on the structure, one must apply the **B**oundary **C**onditions (BC) to the RVE of the volume V. According to [17,23,198], there are several common categories of BC for RVE: KUBC¹, SUBC², PBC³ and MBC⁴. In the case of the periodic volume element, the PBC are usually adopted [1,23,199] to estimate the homogenised behaviour. The periodic stress $\overline{\Sigma}$ leads to the same results as imposed strains \overline{E} . The displacement \overline{u} of periodic cell is associated to every material point \overline{x} and is presented in the following form:

$$\bar{u} = \bar{E}.\,\bar{x} + \bar{\nu} \quad \forall \bar{x} \in \partial V \tag{45}$$

where $\bar{\nu}$ stands for a vector of periodic fluctuations. It corresponds to the periodic part of \bar{u} and possesses the same value for two points that are placed on the opposite side in the RVE of volume V. Hence, the periodic conditions are the next:

$$\bar{\nu}(\bar{x}^{+}) = \bar{\nu}(\bar{x}^{+}) \sigma(\bar{x}^{+}) \cdot \bar{n}^{+} = \sigma(\bar{x}^{-}) \cdot \bar{n}^{-} \quad \bar{n}^{+} = -\bar{n}^{-}$$
(46)

According to the Hill-Mandel theorem, the condition in Eq. 47 is to be complied to provide a valid RVE **[1,16,23,196,199,200]**. Imposed displacement \overline{u} corresponds to macroscopic strain loading E_{ij} . One computes the RVE energy $\langle \overline{\sigma}: \overline{\epsilon} \rangle$ for each of 6 elementary loadings E_{ij} and linear combination (Eq. 48); hence, the stiffness tensor \mathbb{C}^* is computed (Eq. 50 – 51).

$$\langle \overline{\sigma}: \overline{\epsilon} \rangle = \overline{\Sigma}: \overline{E}$$
 (47)

$$\overline{\overline{\Sigma}} = \mathbb{C}^* : \overline{\overline{E}} \tag{48}$$

$$\overline{\overline{E}}: \mathbb{C}^*: \overline{\overline{E}} = <\overline{\sigma}: \overline{\overline{\varepsilon}} > \tag{49}$$

In the case of orthotropic composite material, resulting from orthogonal symmetrical material planes (Figure III-21), the relation between stresses and strains is established with the following homogenised rigidity matrix **[14]**:

¹ Kinetic Unifrom Boundary Conditions

² Static Unifrom Boundary Conditions

³ Periodic Boundary Conditions

⁴ Mixed Boundary Conditions

$$\begin{pmatrix} \Sigma_{11} \\ \Sigma_{22} \\ \Sigma_{33} \\ \sqrt{2}\Sigma_{23} \\ \sqrt{2}\Sigma_{13} \\ \sqrt{2}\Sigma_{12} \end{pmatrix} = \mathbb{C}^* \begin{pmatrix} E_{11} \\ E_{22} \\ E_{33} \\ \sqrt{2}E_{23} \\ \sqrt{2}E_{13} \\ \sqrt{2}E_{13} \\ \sqrt{2}E_{12} \end{pmatrix} = \begin{bmatrix} C_{11}^* & C_{12}^* & C_{13}^* & 0 & 0 & 0 \\ C_{12}^* & C_{22}^* & C_{23}^* & 0 & 0 & 0 \\ C_{13}^* & C_{23}^* & C_{33}^* & 0 & 0 & 0 \\ 0 & 0 & 0 & C_{44}^* & 0 & 0 \\ 0 & 0 & 0 & 0 & C_{55}^* & 0 \\ 0 & 0 & 0 & 0 & 0 & C_{56}^* \end{bmatrix} \begin{pmatrix} E_{11} \\ E_{22} \\ E_{33} \\ \sqrt{2}E_{23} \\ \sqrt{2}E_{13} \\ \sqrt{2}E_{12} \end{pmatrix}$$
(50)

where:

$$C_{11}^{*} = \frac{1 - v_{23}v_{32}}{E_{22}E_{33}\Delta}; \quad C_{22}^{*} = \frac{1 - v_{13}v_{31}}{E_{11}E_{33}\Delta}; \quad C_{33}^{*} = \frac{1 - v_{12}v_{21}}{E_{11}E_{22}\Delta}; \\ C_{12}^{*} = \frac{v_{21} - v_{31}v_{23}}{E_{22}E_{33}\Delta}; \quad C_{13}^{*} = \frac{v_{31} - v_{21}v_{32}}{E_{22}E_{33}\Delta}; \quad C_{23}^{*} = \frac{v_{32} - v_{12}v_{31}}{E_{11}E_{33}\Delta}; \\ C_{44}^{*} = 2G_{23}; \quad C_{55}^{*} = 2G_{13}; \quad C_{66}^{*} = 2G_{12}; \\ \Delta = \frac{1 - v_{12}v_{21} - v_{23}v_{32} - v_{13}v_{31} - 2v_{21}v_{32}v_{13}}{E_{11}E_{22}E_{33}}.$$
(51)

Applying the inversion to the matrix \mathbb{C}^* , the homogenised compliance matrix \mathbb{S}^* is obtained (Eq. 52). It enables the computation of homogenised mechanical properties of the RVE (Figure III-21).

$$\mathbb{S}^{*} = \begin{bmatrix} 1/E_{11} & -\nu_{12}/E_{11} & -\nu_{13}/E_{11} & 0 & 0 & 0\\ -\nu_{21}/E_{22} & 1/E_{22} & -\nu_{23}/E_{22} & 0 & 0 & 0\\ -\nu_{31}/E_{33} & -\nu_{32}/E_{33} & 1/E_{33} & 0 & 0 & 0\\ 0 & 0 & 0 & 1/2G_{23} & 0 & 0\\ 0 & 0 & 0 & 0 & 1/2G_{13} & 0\\ 0 & 0 & 0 & 0 & 0 & 1/2G_{12} \end{bmatrix}$$
(52)

The compliance matrix is symmetric; hence the next equalities are imposed, reducing the number of unknown parameters [14]:

$$\frac{v_{21}}{E_{22}} = \frac{v_{12}}{E_{11}}; \frac{v_{31}}{E_{33}} = \frac{v_{13}}{E_{11}}; \frac{v_{32}}{E_{33}} = \frac{v_{23}}{E_{22}}$$

$$3$$

$$\Sigma_{33}$$

$$\Sigma_{33}$$

$$\Sigma_{31}$$

$$\Sigma_{32}$$

$$\Sigma_{23}$$

$$\Sigma_{2$$

Figure III-21: Representation of six loadings required for the reconstruction of the homogenised matrix [1]

III.3.2. Tow homogenisation: microscale

1

Tow modelling

To recall, tows are formed by randomly distributed fibres linked up with the matrix (Figure III-16). Its mechanical properties are not accessible experimentally; hence, we suggest performing numerical modelling by adopting homogenisation. This requires constituent characteristics. The E-glass fibre properties are selected from the literature and introduced in Table III-3. The matrix properties are already experimentally assessed in Chapter II section II.4.6. The variation of elastic modulus under varying weather conditions is summarised in Figure III-22. Presence of humidity and temperature effects in the matrix will thus provide the environment-related mechanical elastic behaviour of tows.



Figure III-22: Experimental elastic modulus of PA6 as a function of environmental conditions

The RVE of tow is the smallest, yet representative volume, and analogous to the one of UD composite that is widely adopted by researches. Such a cell can be with either periodic fibre arrangement or random. Based on the research works of Abbassi et al. [200], Trias et al. [199] and Dau [23], the choice of RVE is thus oriented towards the periodic cubic structure (Figure III-23). Fibre fraction corresponds to 72.44% as determined during image analysis in section III.2.1. The RVE is, however, created under some hypothesis:

- Possession of perfect and equal theoretical geometry of fibres;
- Perfect contact between fibres and matrix;
- Structured fibre distribution;
- Homogeneous moisture content throughout the matrix;
- Matrix and the experimental neat PA6 are equivalent, though the fabrication process is different.



Figure III-23: Tow RVE for homogenisation at the microscopic scale

Identification of tow properties

In order to identify the homogenised elastic tow properties, it is necessary to apply a unit load. It is represented by traction in 1, 2 and 3 axes and shear in 1-2, 1-3 and 2-3 directions (Figure III-21) **[17,23,199,200]**. The deformed state of the RVE is demonstrated in Figure III-24 for all loading types.



Figure III-24: Von Mises Results of Cast3M simulation of tow for six loading cases E_{ij}

		Ma	trix					Tow				
RH	Т°С	Ε	ν	<i>E</i> ₁₁	<i>E</i> ₂₂	<i>E</i> ₃₃	<i>G</i> ₁₂	G ₁₃	<i>G</i> ₂₃	<i>v</i> ₁₂	ν_{13}	ν_{23}
		MPa	(-)	MPa	MPa	MPa	MPa	MPa	MPa	(-)	(-)	(-)
	-40°C	3773	0.386	55466	16629	16630	4151	4151	5625	0.282	0.282	0.448
	-10°C	3825	0.384	55471	16798	16800	4194	4194	5644	0.282	0.281	0.445
0 %	+23°C	3934	0.415	55727	17570	17571	4211	4211	6043	0.289	0.289	0.498
	+40°C	3831	0.415	55695	17194	17195	4136	4136	5975	0.289	0.289	0.497
	+80°C	1176	0.448	54968	6010	6010	1615	1615	3595	0.309	0.309	0.478
	-40°C	4234	0.353	55477	17980	17982	4553	4553	5684	0.274	0.274	0.407
	-10°C	3923	0.404	55625	17375	17377	4224	4225	5908	0.286	0.286	0.478
50 %	+23°C	1830	0.436	55216	9117	9118	2354	2354	4454	0.299	0.299	0.499
	+40°C	1131	0.449	55011	5854	5854	1556	1556	3618	0.310	0.310	0.495
	+80°C	745	0.457	54827	3959	3958	1066	1066	2938	0.322	0.322	0.495
	-40°C	4940	0.362	55708	20371	20373	4999	4999	6173	0.276	0.276	0.421
	-10°C	2639	0.388	55124	12203	12204	3215	3215	4707	0.285	0.285	0.429
85 %	+23°C	1135	0.448	54998	5858	5858	1562	1562	3603	0.310	0.310	0.491
	+40°C	764	0.457	54845	4063	4062	1091	1091	2990	0.322	0.322	0.497
	+80°C	516	0.461	54655	2791	2788	757	757	2359	0.337	0.337	0.492

Table III-4: Elastic properties of PA6 GF tow obtained with homogenisation method at different environmental conditions

The computation of the inversed homogenised stiffness and, thus, the compliance matrix provides the mechanical properties of tows. Characterised matrix at different RH levels and temperatures enables likewise the complete estimation of tow behaviour. All the input matrix parameters and resulting output tow properties are

outlined in Table III-4. These results subsequently serve as the input for the identification of homogenised elastic composite material behaviour (section III.3.3).

Analysing Table III-4, one can track the analogy between mechanical behaviour of tows and UD composites that occurs due to a similar transverse isotropic structure. The longitudinal elastic modulus remains quasiunchanged for all environmental conditions by reason of fibre domination, whereas transversal and out-of-plane moduli are significantly smaller and tend to diminish with the increase of temperature/humidity. The same trend concerns the shear moduli. It is due to the matrix impact, hence, coherent with its modulus. Poisson's ratios also undergo relevant changes. Consequently, exposed to varying environment composite material is not only affected by the altering matrix behaviour, but also by that of the tows. Similar observations and conclusions are made by Dau **[23]**.

III.3.3. Composite homogenisation: mesoscale

The second homogenisation on the mesoscale is required in order to estimate all the mechanical elastic properties of the composite PA6 GF. Like a real material, its RVE is composed of woven fabric with twill weave 2×2 (Figure III-25 a) and matrix (Figure III-25 b). The matrix properties, detailed in Table III-4, are employed to characterise the composite at each coupled environmental condition. As for the tows, the formerly computed properties are employed (Table III-4). Analogously to the homogenisation at the micro-scale, six unit loads (traction and shear presented in Figure III-21) are applied in order to obtain homogenised stiffness matrix of the RVE. Its inversion determines the compliance matrix with sought elastic properties (Eq. 52).

The RVE is modelled and homogenised under the hypotheses, similar to those described in the previous section:

- Perfect and equal theoretical geometry of tows;
- Equal vertical and horizontal distances between tows;
- Perfect contact between tows and matrix;
- Homogeneous moisture content throughout the matrix and tows;
- Matrix and the experimental neat PA6 are equivalent, though the fabrication process is different;
- Behaviour of glass fibre does not change with temperature.



Figure III-25: Overview of RVE homogenisation. The notations stand for: a) homogenised fabric; b) matrix and c) composite ply composed of the fabric and the matrix

The numerical simulations allow the identification of the homogenised out-of-plane properties of the composite material, demanded for computations of bolted joint parameters. Nonetheless, a prior validation of homogenised RVE model is essential. It is performed by comparing in-plane numerical to experimental composite properties from Chapter II section II.4.4.

Validation of numerical elastic in-plane mechanical properties

The homogenised RVE of the composite PA6 GF is to be validated prior to any interpretation and use of out-of-plane properties. Experimental campaign on 0°, 90°, 45°-oriented specimens (Chapter II section II.4.4), scientific articles and theses on the same or similar composite materials enable the comparison of numerical and experimental results.







Figure III-27: Experimental and numerical transversal elastic moduli of PA6 GF versus RH and varying temperature

Figure III-26 and Figure III-27 illustrate the relation between longitudinal/transversal elastic modulus and testing environmental conditions (summarised in Table III-5 at the end of the section), obtained from the

mechanical testing campaign in Chapter II. To recall, 0° and 90°-fibre orientation is particular and provides the fibre-dominated properties. Owing to the homogenisation at the micro-scale, the material is rather tow-dominated in the present case. Hence, both fibres (in a significant way) and matrix (in a less significant way) are engaged in mechanical performance. The following observations can be pointed out:

- Numerical longitudinal and transversal moduli are quasi-equivalent, except the small variations at -40°C;
- Composite moduli follow the same tendency as the tow longitudinal modulus, regardless of the RH level;
- Numerical results are not entirely coherent with the experimental results. The former possess a more noticeable shift towards lower moduli values. It is also slightly marked for experimental E_{11} , but not valid for E_{22} ;
- Most remarkable difference between numerical and experimental data at RH 50% is noticed at +23°C and +80°C, whereas at -40° it is decayed;
- Homogenised values are lower than the experimental.

Shear properties of the composite material are expected to be rather matrix-dependent; however, the influence of tows is also present. Experiments, carried out at all studied environmental conditions, allow the comparison to homogenised properties at each temperature and moisture content due to exposure to three RH levels. From Figure III-28 one can note the following remarks:

- Both numerical and experimental shear moduli possess a strong relation to the experimentally obtained matrix properties (Figure III-22). The drastic material changes are caused by decreasing glass transition temperature of PA6 as outlined in Chapter II section II.2.6;
- Experimental properties are greater than numerical at -40°C and -10°C; though, they become slightly lower at ≥+23°C;
- Most important discrepancy is noted at -40°C for all RH and at -10°C for RH 0% and 85%;
- Despite some differences at negative temperatures, the homogenised elastic properties are consistent with the experimental.



Figure III-28: Experimental and numerical shear moduli of PA6 GF as a function of RH and varying temperature

Similar conclusions for longitudinal, transversal and shear behaviour are also made by Dau [23] for PA66 GF tested at the ambient temperature and three RH. The difference between experimental and modelled behaviour may arise from the moisture content in matrix and tows. Considering the shear at -40°C and tow-dependent experimental values, homogeneous moisture distribution may be a strong assumption for numerical properties. Besides, the approach for the computation of tow properties may be not entirely appropriated due to the choice of fibre positioning. Thus, the tow RVE with the only fibre in the middle may lead to different results as compared to an RVE with a number of randomly distributed fibres [17]. The other proposed hypothesis consists in the mechanical behaviour of thermoplastic polymer PA6. The fabrication process of the neat PA6 specimens and the composite, where this resin is used as the matrix, is different. Consequently, their theoretical mechanical properties are not expected to be identical. The use of the neat polymer properties for the composite matrix is, therefore, not totally defensible. One can also suggest that the discrepancy between experimental numerical values decreases in the vicinity of T_g for each RH level: T_g is \approx -1°C (\approx -10°C in [55,58,96]) for RH 85%, +23°C for RH 50% and +55.8°C (+60°C from [163]). For instance, it can be observed at the testing temperature of -10°C.

Nevertheless, the overall comparison of elastic homogenised and experimental properties is satisfying, thus, favouring the estimation of out-of-plane numerical properties under thermal and humid impacts.

Identification of numerical elastic out-of-plane properties

Characterisation of out-of-plane elastic mechanical behaviour is the main target of numerical modelling in the present project. Following the successive double-scale homogenisation, the range of in-plane numerical properties is found to be satisfactory in comparison to experimental ones. As a consequence, one can predict the elastic modulus E_{33} of the composite as a function of humid and thermal conditions (Figure III-29 and Table III-5). The complexity of the experimental campaign makes it difficult to compare out-of-plane homogenised to experimental results. However, Hamonou [1] proposed other methods and results for unconditioned composite PA6 GF.

As one can see from demonstrated results, the out-of-plane elastic modulus obeys the identical trend as the homogenised in-plane shear (Figure III-28) and, thus, the matrix PA6 (Figure III-22). What is interesting to note, such behaviour is valid for all RH. The principal reason is the obvious matrix-dependency of out-of-plane behaviour.

Consequently, the hypothesis of homogeneous moisture content throughout the matrix has a direct effect on the modulus E_{33} . As expected, the modulus at RH 85% and all the temperatures is lower than at RH 0% and 50% apart from -40°C. Similarly, the elastic modulus at RH 50% is higher than that at RH 0% at negative temperatures. We presume it to be linked to the presence of moisture in the composite that may have a particular effect at negative temperatures. The sudden reduction of E_{33} is also visible in the vicinity of the glass transition temperature for each relative humidity.

Confirmation or rejection of applied double-scale homogenisation and computed out-of-plane properties can be performed while comparing the present results to those, obtained with the other approaches. Hamonou [1] used conventional non-destructive ultrasonic testing, Linear Variable Differential Transformer (LVDT) and extended Finite Element Method (x-FEM) for non-conditioned PA6 GF at ambient temperature. The results are represented in Figure III-29 for direct comparison. Due to prolonged storage under uncontrolled conditions in a laboratory, one can suppose that the material has the moisture content similar to the composite, conditioned between RH 55% and 70% (usual RH in the region). Following this assumption, the homogenised properties at +23°C and RH 50% and 85% seem to be coherent to those obtained by Hamonou. In addition, analogous results are achieved by Dau [23] for PA66 GF at different RH levels.

Eventually, the importance of humidity and temperature on both in-plane and out-of-plane mechanical elastic behaviour of the composite PA6 GF is confirmed. The moisture content is of absolute importance for out-of-

plane elastic moduli; hence, it plays a significant role in the durability of bolted joints, as demonstrated in the following Chapter IV.



Figure III-29: Numerical out-of-plane elastic moduli E_{33} of PA6 GF under environmental conditions

		Ma	trix		Composite							
RH	Т℃	E	ν	<i>E</i> ₁₁	<i>E</i> ₂₂	<i>E</i> ₃₃	<i>G</i> ₁₂	<i>G</i> ₁₃	G ₂₃	<i>v</i> ₁₂	ν ₁₃	ν_{23}
		MPa	(-)	MPa	MPa	MPa	MPa	MPa	MPa	(-)	(-)	(-)
	-40°C	3773	0.386	24345	24344	10830	3355	1848	1848	0.146	0.419	0.419
	-10°C	3825	0.384	24417	24416	10911	3373	1870	1870	0.147	0.417	0.417
0 %	+23°C	3934	0.415	24903	24902	11953	3575	1904	1904	0.157	0.459	0.459
	+40°C	3831	0.415	24727	24726	11702	3525	1862	1862	0.155	0.459	0.459
	+80°C	1176	0.448	19446	19444	4799	1910	672	672	0.083	0.527	0.527
	-40°C	4234	0.353	24884	24883	11271	3465	2052	2052	0.147	0.379	0.379
	-10°C	3923	0.404	24759	24758	11615	3511	1902	1902	0.154	0.443	0.443
50 %	+23°C	1830	0.436	21018	21017	6793	2447	991	992	0.104	0.507	0.507
	+40°C	1131	0.449	19382	19381	4771	1909	650	650	0.082	0.539	0.539
	+80°C	745	0.457	18266	18264	3477	1506	449	449	0.071	0.565	0.565
	-40°C	4940	0.362	26025	26024	12968	3806	2322	2322	0.162	0.386	0.386
	-10°C	2639	0.388	22302	22301	8030	2726	1364	1365	0.119	0.426	0.426
85 %	+23°C	1135	0.448	19380	19378	4757	1903	652	652	0.082	0.537	0.537
	+40°C	764	0.457	18335	18333	3559	1534	459	459	0.071	0.564	0.564
	+80°C	516	0.461	17416	17414	2587	1186	323	323	0.067	0.582	0.582

Table III-5: Elastic properties of the composite obtained with homogenisation methods at different environmental conditions

III.3.4. Conclusions

The complex and inappropriate reviewed experimental methods lead to the exploration of other techniques determining composite out-of-plane behaviour and the elastic modulus in particular. The latter is essential for the proper dimensioning of bolted joints for industrial applications. We propose a methodology that is based on homogenisation and finite element methods with the use of fibre and matrix properties to avoid costly and time-

consuming experimental characterisation. The microscale and mesoscale RVEs, generated in accordance with the real composite geometry, serve for double-scale homogenisation. Initially, the homogenised elastic properties of tows are computed on the periodic cubic structure at the micro-scale. Then they are integrated into the full-scale RVE of woven 2×2 structure for the computation of the whole range of mechanical properties using Cast3M-CEA.

Obtained results confirm the significant impact of the humid environment on the tow behaviour, in-plane and out-of-plane composite properties. The numerical outcome is in agreement with the experimental campaign, except for negative temperatures, where the experimental data is superior to homogenised ones. This is supposed to be related, firstly, to the strong assumption of the uniform moisture distribution throughout the composite and, secondly, to the dissimilar behaviour of composite with high moisture content at negative temperatures that are well below the material glass transition. The other two hypotheses consist in: 1) dissimilarity of the matrix and the used neat resin properties due to the fabrication process; 2) the choice of periodic tow RVE may result in over/underrated stiffness of tows for the conditions below the glass transition temperature. These hypotheses are yet to be verified. Nevertheless, the out-of-plane characteristics of the composite are demonstrated to be directly affected by the matrix as a result of revealed similar changing behaviour in relation to humidity and temperature.

Out-of-plane elastic moduli are subsequently employed for computation of composite compliance and analysis of bolted joint behaviour over time as presented in Chapter IV.

CHAPTER IV

PERFORMANCE OF PRELOADED COMPOSITE BOLTED JOINTS

Performance of a bolted structure is dependent on a number of parameters, related to elementary constituents of bolted joints, type and position of acting loadings, working environment, etc. Provided that the composite materials are used, the impact of mechanical behaviour of assembled members over time may be intensified. This can lead to the overall degradation of mechanical performance of bolted connections affecting structural integrity. One of the principal reasons is an important weakening of preload, applied to bolted joints. Furthermore, the reduction may be aggravated by the environmental conditions through the lower resistance to creep. Hence, an experimental evaluation of composite bolted joints, subjected to preload, is proposed. The relations between mechanical, experimental and numerical properties of the thermoplastic composite and its impact on the joint behaviour are demonstrated with the subsequent analysis. Member compliance is also investigated for different geometrical and environmental parameters.

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	CHARACTERISATION OF COMPOSITE MATERIAL COMPLIANCE

IV.1. **CHARACTERISATION COMPOSITE MATERIAL** OF **COMPLIANCE**

Composite material stiffness has a significant impact on the capacity of joints to maintain the applied preload over time. The member compliance is, therefore, dependant on several parameters, related to the joint geometry and material mechanical characteristics. This section provides the evaluation of member compliance as a function of environmental conditions using the standard approach, developed for metallic materials with the deformation cone angle of 45°, and an elsewhere proposed method by using a true angle for a given member diameter and thickness.

IV.1.1.Introduction

The particularity of bolted joints consists in the ability to postpone the failure to a required extent. It is possible through the application of pretension force F_0^b to bolt. Hence, as demonstrated in section I.2.2.3 and recalled in Figure IV-1, the resultant force in the bolt F_b is following [160]:

$$F_b = F_0^b + \lambda F_{ext} \tag{54}$$

Developed analytical models for dimensioning and computation of bolted metal connections [159] describe the joint as a combined spring system with the restricted to a conical pressure zone (on the right in Figure IV-2). When the preload is applied, the tension force in a bolt is balanced by compressive forces occurring in joined parts as presented in Figure IV-2. The system is, thus, composed of bolt and member elastic compliances due to an assumption of the elastic behaviour in the vicinity of bolt axis [1]. A model with the spring system relates the applied preload, the external load and the compliance of the bolt and parts. The latter is associated with the filtering coefficient λ , which represents the slope prior to joint failure (Figure IV-1). To recall, the equation is expressed as follows:

$$\lambda = \beta \frac{\delta_p}{\delta_b + \delta_p} \tag{55}$$

where β stands for the load introduction factor, $0 \ge \beta \ge 1$.



Figure IV-2: Mechanical model of a bolted joint [1,160]

Stiffness of assembled materials and bolts E_{zz} is one of the significant parameters for good performance of bolted joints. Nevertheless, an equivalent cross-section of joined parts and their thickness are as well important as can be seen from the following equation, recalled from section I.2.2.3:

$$\delta_p = \frac{1}{A_{eq}} \sum_{i=1}^{l=n} \frac{L_{pi}}{E_{zzi}}$$
(56)

There are three cases that define the equivalent cross-section of metallic parts. They are related to the compressed and deformed zone with the cone angle equal to 45° **[159,160]**:

• Deformation cone is hardly present $D_p \leq d_w$ (Figure IV-3 a)

$$A_{eq} = \frac{\pi}{4} \left(D_p^2 - D_h^2 \right) \tag{57}$$

• Deformation cone is truncated $d_w < D_p < d_w + L_p$ (Figure IV-3 b)

$$A_{eq} = \frac{\pi}{4} \left(d_w^2 - D_h^2 \right) + \frac{\pi}{8} d_w \left(D_p - d_w \right) (X+2) X; \quad X = \sqrt[3]{\frac{L_p d_w}{D_p^2}}$$
(58)

• Deformation cone is entirely included $D_p \ge d_w + L_p$ (Figure IV-3 c)



Figure IV-3: Schematic illustration of the deformation cone as a function of member diameter. d_w is a diameter of the washer head, D_h is a clearance hole diameter, D_p is a diameter of the clamped members, D_c is a diameter of the deformation cone at the interface surface of clamped members [1]

Unlike the metallic materials, the orthotropic composites have heterogeneous structure with different sizes of RVEs resulting in potentially different deformation cone. Hence, the compliance of composite parts within a bolted joint is a relatively challenging issue. Hamonou [1] proposed three approaches for the identification of compliance of composite materials with varying member diameter: analytical, based on the energetic criterion developed in [159], numerical [201] and experimental. The analytical method consists in the energy equivalency between a three-dimensional and an equivalent one-dimensional environment with the restriction to its out-of-plane component. The total compliance of the centred, axially loaded bolted joint is defined as:

$$\delta_{total} = \delta_b + \delta_{p1} + \delta_{p2} = \sum_{i=1}^{n=3} \frac{1}{F^2} \int_{V_{ri}} Tr[\bar{\sigma} \ \bar{\varepsilon}] dV_{ri} , \qquad Tr[\bar{\sigma} \ \bar{\varepsilon}] \cong \sigma_{33} \varepsilon_{33}$$
(60)

where V_{ri} is the volume of interest. Numerical identification is implemented in Cast3M-CAO with the use of energy equivalence as for analytical computation. Results of the numerical finite element model demonstrate the discrepancy from the analytical output of CETIM-Cobra V6 (Figure IV-4). It originates from used in numerical modelling empirical formula of cone angle (Eq. 61), which is dependent on the compressed length of members L_p . All the data is, however, computed for the only value of elastic modulus E_{33} .



Figure IV-4: Comparison of analytical (COBRA V6) and numerical (Cast3M) results of PA66 T700 compliance [1]. "Souplesse" stands for compliance, "diamètre" is diameter

An experimental approach, proposed in [1], is based on the LVDT measurements during compressive testing of thermoplastic composites. A similar tendency to stabilisation of compliance starting from the diameter of 28 mm is explained by the entire inclusion of deformation cone in the material PA66 T700 (Figure IV-5). According to Hamonou [1], a strong variation of PA6 GF compliance can be explained by a more ductile behaviour and another ration of d_w/RVE . The volume of fibres in PA6 GF is not necessarily the same in the specimens of all the tested diameters. This phenomenon is less observable in PA66 T700 by reason that its RVE size is twice smaller than that of PA6 GF. Despite different approaches, the correlation between analytical, numerical and experimental results is assumed to be satisfying.



Figure IV-5: Experimental results of PA66 T700 and PA6 GF compliance for 6 mm thickness [1]. "Souplesse" stands for compliance, "diamètre" is diameter

As seen from presented figures, neither of the proposed methods could be selected as credible for the composite material due to:

- Heterogeneous structure;
- Varying RVE;
- Rigid/ductile behaviour of the matrix;
- Unknown angle of pressure cone in relation to the size of the RVE;

Strong interrelation between compliance, member diameter and washer head diameter.

Nevertheless, the application of the analytical approach with the standard for metals angle $\varphi = 45^{\circ}$ and parametrically computed by [1] angle φ may demonstrate the ultimate values of composite compliance. The experimental approach is not manifested to be reliable due to strong data variation for PA6 GF. The analytical compliance of PA6 GF is presented in the following section.

IV.1.2. Determination of compliance

The aim of this section is to estimate one of the parameters defining mechanical behaviour of bolted composite joints – the compliance of PA6 GF. The interest lies in evaluating compliance as a function of studied environmental conditions by using the above-mentioned analytical approach. The numerically computed out-of-plane elastic modulus E_{zz} is employed here (data is taken from section III.3.3 Table III-5).

The choice of specimen geometry is determined by its use in the experimental campaigns in Chapters II and IV (section IV.2). The selected specimen is such that the deformed volume, which maintains the bolt prestress is completely introduced (Table IV-1) enabling the use of the case C (Eq. 59) for computation of equivalent cross-section as demonstrated in Figure IV-3. These specimens are graphically presented in section II.1.2.2 (Figure II-9 d, e and f). The thickness is 2 mm, 4 mm and 6 mm.

$D_h = 13 mm$	$d_w = 17.23 \ mm$	$D_p = 36 mm$
		1 1 1

Та	ble	IV	-1:	Geometrical	parameters of	of assem	bled	members
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Analytical approach (VDI recommendation)

According to the case C (Eq. 59) and Eq. 56, the composite compliance δ_p is computed for three total specimen thicknesses: 4 mm when 2 specimens of 2 mm are joined, 8 mm for 2 specimens of 4 mm and 12 mm for 2 specimens of 6 mm. This choice is justified by the exact geometry used in section IV.2.

One can notice from Figure IV-6 that environmental conditions, i.e. increase in RH and temperature lead to the rise of compliance for every studied thickness. Such a tendency is, apparently, associated with the out-ofplane elastic modulus variation (Figure III-29). Ductile composite behaviour contributes to higher compliance for whichever thickness (for instance, at +80°C/RH 85%) in comparison with the rigid material (at -40°C). In addition, member thickness can be considered as an equally significant parameter in the case of ductile material state ($T \ge T_g$) by reason of very similar values observed in the rigid state at -40°C ($T < T_g$ for all specimens). The results of compliance are summarised in Table IV-2 at the end of the present section.

A principal disadvantage of the used method is that the cone angle is expected to be 45° once compressive deformation occurs. Although such formulation of a zone of interest is applicable to isotropic materials, the composite material does not behave similarly. Hence, the zone of interest may be bordered by the other angles, as suggested by Hamonou [1]. Despite the mentioned drawbacks, the computed values are expected to define the limits of compliance for studied geometries at given environmental conditions as demonstrated in Figure IV-4.

A relation of the angle φ and a member diameter is proposed in [159] (Eq. 61) for bolted joints and studied by Hamonou [1] for 6 mm and 12 mm member thicknesses. This equation is empirical, has no physical background and integrates the geometry suitable for metallic materials due to the isotropic structure.

$$tan(\varphi) = 0.362 + 0.032 \ln\left(\frac{L_p}{2}\right) + 0.153 \ln(D_p)$$
(61)



Figure IV-6: Prediction of composite compliance versus part geometry and environmental conditions

Hamonou [1] identified new coefficients for the model in Eq. 61 by minimising between numerical [201] and analytical [1] (Eq. 63) results for the composite material PA66 T700 using the least-squares method. The results of the angle φ are illustrated in Figure IV-7 as a function of a part diameter. The angle values for $D_p = 36 \text{ mm}$ (Eq. 62) would contribute to the definition of compliance range on condition that the total thickness of assembled parts is either 12 mm or 6 mm.

 δ_p

$$\varphi = 22.01^{\circ}, \ L_{p} = 12 \ mm \qquad (62)$$

$$= \frac{2ln\left(\frac{(d_{w} + D_{h})(L_{p}tan\varphi + d_{w} - D_{h})}{(d_{w} - D_{h})(L_{p}tan\varphi + d_{w} + D_{h})}\right)}{D_{h}tan\varphi E_{zz}\pi}$$



Figure IV-7: Angle φ as a function of member diameter ("diamètre des pieces") for 12 mm thickness [1]

Computed results for $\varphi = 22.01^{\circ}$ according to Eq. 63 are demonstrated in Figure IV-8 in relation to environmental conditions, in Figure IV-9 in relation to the elastic modulus E_{33} and compared to $\varphi = 45^{\circ}$. The outcome for $L_p = 6 \, mm$ is equally plotted and illustrated in Appendix F. All results are eventually outlined in Table IV-2.


Figure IV-8: Composite compliance as a function of environmental conditions and cone angle φ for 12 mm thickness



Figure IV-9: Composite compliance as a function of elastic modulus and cone angle φ for 12 mm thickness

From Figure IV-8, one can see the identical tendency of composite compliance growth for both cone angles. Whatever the temperature and RH, the values, computed for $\varphi = 22.01^{\circ}$, seem to define the upper limit of potential part compliance, which is comparable to results in [1]. What is more, the increasing difference between results at 22.01° and 45° correspond to reducing composite stiffness. Such behaviour is apparent in Figure IV-9. Most important changes occur till 8000 MPa of E_{zz} for RH 50% and 85%. Once again, the ductile state of PA6 GF is strongly related to the glass transition temperature T_g as discussed in sections I.1.3.2 and I.1.4.2. Both graphs validate that the relevance of the cone angle significantly decreases towards greater elastic modulus, or stiffer composite with the given thickness and diameter.

Considering the fact that the applied approach (Eq. 61) is initially elaborated for isotropic materials, the computed results for the orthotropic composite remain approximated.

Chapter IV	Performance o	f preloaded	composite	bolted	joints
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								T	
		F		δ_{p} , $ imes$ 10	$\varphi = 22.01^{\circ}$	$arphi=20.91^\circ$			
RH T°C	Т°С		6 mm	6 mm	4 mm	2 mm	4 mm	6 mm	6 mm
		WII a	2 sp.	1 sp.	2 sp.	2 sp.	1 sp.	2 sp.	1 sp.
	-40°C	10830	4.263	3.008	3.527	2.314	2.314	5.959	3.622
	-10°C	10911	4.231	2.986	3.501	2.296	2.296	5.915	3.595
0 %	+23°C	11953	3.862	2.725	3.195	2.096	2.096	5.399	3.282
	+40°C	11702	3.945	2.784	3.264	2.141	2.141	5.515	3.352
	+80°C	4799	9.620	6.788	7.959	5.221	5.221	13.448	8.174
	-40°C	11271	4.096	2.890	3.389	2.223	2.223	5.726	3.480
50 %	-10°C	11615	3.975	2.805	3.288	2.157	2.157	5.556	3.377
	+23°C	6793	6.796	4.796	5.623	3.689	3.689	9.501	5.775
	+40°C	4771	9.677	6.828	8.006	5.252	5.252	13.527	8.222
	+80°C	3477	13.278	9.369	10.985	7.206	7.206	18.562	11.282
	-40°C	12968	3.560	2.512	2.945	1.932	1.932	4.977	3.025
	-10°C	8030	5.749	4.057	4.757	3.120	3.120	8.037	4.885
85 %	+23°C	4757	9.705	6.848	8.029	5.267	5.267	13.567	8.246
	+40°C	3559	12.972	9.153	10.732	7.040	7.040	18.134	11.022
	+80°C	2587	17.846	12.593	14.764	9.686	9.686	24.947	15.163

Table IV-2: Composite compliance at different environmental conditions, part geometry and angle φ

Numerical approach

A numerical approach is employed for more rigorous computation of part compliance. Similarly to the recommended by VDI 2230 analytical method, this approach is also based on the energy equivalency detailed in section IV.1.1. The volume of interest V_i is bordered by the iso-zero values of volumetric deformation energy reduced to the out-of-plane component [201]:

$$E_{33} = \frac{1}{2}\sigma_{33}\varepsilon_{33} \tag{64}$$

Hence, by applying the finite element model and nine mechanical properties of the composite, one can compute the compliance of assembled parts:



Figure IV-10: Geometry and boundary conditions of numerical model [1]

The geometry of modelled specimen is demonstrated in Figure IV-10. It has been developed and employed by Gornet [201] and Hamonou [1]. In the present work the next conditions are used: $d_w = 18 \text{ mm}$; $D_h = 13 \text{ mm}$; $D_p = 36 \text{ mm}$; $L_p = 4$, 8 and 12 mm (Figure IV-10). The applied pressure is 50 MPa. The

computation is realised in Cast3M-CEA. The occurring out-of-plane deformations and stresses are illustrated in Figure IV-11.



Figure IV-11: Numerical simulations with the finite element method in Cast3M-CEA: out-of-plane deformation ε_{zz} is presented in a) for 4 mm thickness and b) for 12 mm thickness; out-of-plane stress σ_{zz} is presented in c) for 4 mm thickness and d) for 12 mm thickness



Figure IV-12: Numerically computed composite compliance as a function of part geometry and environmental conditions

The computed results of compliance of the composite material PA6 GF are summarised in Table IV-3. The numbers in the parentheses represent the difference between analytical and numerical values. It is proposed that, depending on the member thickness, the inaccuracy in analytical computations can be rectified without the need to identify the angle of the deformation cone. One can note that the difference between analytical and numerical composite compliance has a strong tendency to increase for thicker clamped members. Less pronounced

changes of δ_p can be seen for higher material ductility. However, none of the approaches considers the presence of an interface surface. For this reason, the suggested numerical approach should be expanded in order to introduce one or more interface surfaces between clamped parts. In addition, the use of different materials may be of interest for multi-material bolted joints.

F		F	δ_{p} ,× 10	$^{-6} mm/N: ~ o$	$arphi=45^\circ$	$\delta_{p} \times 10^{-6} \ mm/N$: numerical			
RH	Т℃	L MPa	6 mm	4 mm	2 mm	6 mm	4 mm	2 mm	
		1 111 a	2 sp.	2 sp.	2 sp.	2 sp.	2 sp.	2 sp.	
	-40°C	10830	4.263	3.527	2.314	5.579 (23.6%)	4.268 (17.4%)	2.429 (4.7%)	
	-10°C	10911	4.231	3.501	2.296	5.537 (23.6%)	4.236 (17.4%)	2.411 (4.8%)	
0 %	+23°C	11953	3.862	3.195	2.096	5.047 (23.5%)	3.838 (16.8%)	2.165 (3.2%)	
	+40°C	11702	3.945	3.264	2.141	5.157 (23.5%)	3.924 (16.8%)	2.215 (3.3%)	
	+80°C	4799	9.620	7.959	5.221	12.79 (24.8%)	.8%) 9.811 (18.9%) 5.615		
i									
	-40°C	11271	4.096	3.389	2.223	5.358 (23.6%)	4.111 (17.6%)	2.351 (5.4%)	
-10°	-10°C	11615	3.975	3.288	2.157	5.198 (23.5%)	3.962 (17.0%)	2.243 (3.8%)	
50 %	+23°C	6793	6.796	5.623	3.689	8.984 (24.4%)	6.866 (18.1%)	3.908 (5.6%)	
	+40°C	4771	9.677	8.006	5.252	12.91 (25.0%)	9.892 (19.1%)	5.656 (7.1%)	
	+80°C	3477	13.278	10.985	7.206	17.82 (25.5%)	13.67 (19.6%)	7.843 (8.1%)	
	-40°C	12968	3.560	2.945	1.932	4.656 (23.5%)	3.562 (17.3%)	2.027 (4.7%)	
	-10°C	8030	5.749	4.757	3.120	7.526 (23.6%)	5.78 (17.7%)	3.311 (5.8%)	
85 %	+23°C	4757	9.705	8.029	5.267	12.94 (25%)	9.917 (19.0%)	5.672 (7.1%)	
	+40°C	3559	12.972	10.732	7.040	15.92 (18.5%)	12.51 (14.2%)	7.405 (4.9%)	
	+80°C	2587	17.846	14.764	9.686	21.86 (18.3%)	17.23 (14.3%)	10.25 (5.5%)	

Table IV-3: Numerical and analytical results of composite compliance. Numbers in parentheses correspond to the difference between numerical and analytical results

IV.1.3. Conclusions

The importance of stiffness of bolted joints constituents is discussed in the section. The attention is given to the compliance of bolted structures made with the composite material PA6 GF. The use of the method, initially developed for metal joint ($\varphi = 45^{\circ}$), is incorrect for the composite joint. Only a numerical homogenisation approach can ensure the valid results for the orthotropic material. Nevertheless, it is proposed to employ two fictive values of the cone angle to estimate the range of member compliance at different environmental conditions. The first angle is 45°, used for isotropic materials (classically metals), and the second is 22.01° for the diameter 36 mm from the literature review. Results show that the most important changes in compliance occur at relatively low values of the out-of-plane elastic modulus, computed numerically in Chapter III. It is assumed to be related to the ductile material behaviour. What is more, as presented in the following section, the observed changes of member compliance, caused by the environment and member geometry, enable forming an opinion on the joint capacity to maintain the preload over time.

Nevertheless, due to the heterogeneous material structure, obtained results of member compliance are revised with a numerical method. Obtained compliance values are demonstrated to be somewhat higher than those computed with the analytical approach. The strong tendency to increasing difference between both methods is noted for bigger member thickness. For industrial applications and low calculating cost, the analytical results are proposed to be adjusted for orthotropic composite materials by using the computed difference between values as a function of material thickness. However, the numerical model necessitates the extension for the multi-material joints with a few interface surfaces.

IV.2. EXPERIMENTAL CHARACTERISATION OF PRELOAD EVOLUTION

The interest of this experimental campaign consists in the evaluation of preload loss after tightening of bolted assembly of composite materials. The importance of the study is to reveal whether ductility of the composite under certain environmental conditions in combination with heterogeneous structure may lead to more rapid bolt loosening. The standard for bolted joint dimensioning is theoretically developed for entirely metallic joints and does not include the environmental effects, except thermal expansion. Hence, the characteristics of a bolted composite joint are not evident to determine for proper mechanical behaviour. Some of the required parameters are obtained and analysed during the experiments with the consequent comparison to the simultaneously tested bolted metallic joints. They come from the impact of environment and member geometry. To avoid bearing damage of composite specimens, the preload value is determined for RH 85% condition. Bolts are calibrated till 60 kN in order to eliminate unwanted effects from gauge instrumentation and obtain equations describing bolt behaviour in traction.

IV.2.1. Methodology

As already demonstrated in the literature review in Chapter I, the loss of mechanical properties of assembled parts has an unfavourable effect on the durability of prestressed bolted joints. According to results from Chapter III and section IV.1, mechanical behaviour of the composite PA6 GF is indeed environment-dependent; hence, the elastic parameters undergo an important change. The sudden drop of mechanical properties and the increase of creep deformation emphasise the viscous nature of the thermoplastic material. This leads to the decrease of capacity to sustain a high level of preload, also required to ensure fatigue endurance of fasteners [159]. Hence, an experimental campaign is launched in order to evaluate the preload loss over time when joints are exposed to regulated environmental conditions.



Figure IV-13: Causes of bolt loosening1

The loss of applied pretension in a bolt can be characterised by two distinct phases: spontaneous loss and long-term loss, induced by different factors as depicted in Figure IV-13. Heistermann [150] similarly observed such a tendency for metallic members with galvanised surfaces. Considering such behaviour, the test duration should be sufficiently long to represent joint performance. The joints are prestressed twice during one test according to the next steps:

• First loading (torque) is applied until the maximum defined force (section IV.2.2.2);

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- Test runs for 7 10 days due to limited available time;
- Second loading (torque) is subsequently applied until the same level without unscrewing the nuts;
- Test runs for 7-10 days.

All the gauge data is recorded from the beginning of the experiment by QuantumX MX 1615 and MX 1615B of a trademark HBM®. The experimental set-up is placed into the controlled in temperature and relative humidity climatic chamber (Figure II-14) after the first and second tightening. The experimental configuration is demonstrated in Figure IV-14. One can see 5 configurations of bolted joints tested simultaneously, where each of them is repeated three times for the repeatability of results. Three entirely metallic joints (on the right in Figure IV-14) are used to compare and analyse the response of composite joints; its member is composed of 4 thick washers. What is more, a free bolt is equally placed in the climatic chamber. It is necessary in order to eliminate unwanted possible deformations of the strain gauges and the adhesive caused by environmental conditions.

In relation to joined members, not only their mechanical behaviour is important, but also the number of contact faces **[159]**. Thus, it is of great interest to analyse this effect in more detail by using different composite thicknesses. Extra information regarding the specimens can be found in section IV.2.2.1. The preparation of bolts prior to testing is outlined in section IV.2.2.3 and the definition of preload level – in section IV.2.2.2.



Figure IV-14: Experimental setup for preload characterisation

IV.2.2. Preparation stage

IV.2.2.1. Definition of test matrix

The composite circular specimens with a centred hole are used in the experimental campaign on preloaded bolted joints. Their geometry has been already introduced in Chapter II section II.1.2.2 in Figure II-9 (d, e f). The use of the smaller diameter, 18 mm, is unjustifiable by reason of not included full RVE. Hence, the accurate material response cannot be ensured when the force is applied. It is related to the irregular stiff and ductile composite areas in the compression zone.

Type of composite	Specimen	Spe	Environmental				
material	external diameter	clearance hole	one specimen	total	number of specimens	conditions	
PA6 GF	36 mm		2	4	2		
		13 mm	4	4	1	+23°C/RH 50%	
			4	8	2	+23°C/RH 85%	
			6	12	2		

Table IV-4: Test matrix for experimental characterisation of preload

The test matrix is presented in Table IV-4. The different specimen thicknesses are compensated by the washers of three geometries and the metallic plate of 20 mm thickness (Figure IV-14). Eventually, the working bolt length is constant and equal to 42 mm. The use of 1 or 2 specimens of the equal total thickness is necessary in order to compare the impact of the interface surface between the clamped members. As for environmental conditions, RH 50% and RH 85% are selected due to the limitations of the climatic chamber making it difficult to maintain the condition RH 0% for a prolonged period of time. All the used specimens are initially conditioned according to the protocol, proposed in Chapter II section II.2. Unconditioned specimens, not presented in Table IV-4, are used to set up the testing procedure.

IV.2.2.2. Determination of preload

The composite material PA6 GF, aged at RH 85%, undergoes the biggest mechanical deformation comparing to RH 0% and 50% as experimentally examined and presented in Chapter II section II.4.4. It occurs due to the viscous nature of the composite when the in-service temperature exceeds the glass transition T_g . Such condition can be, therefore, considered as the most critical not only for in-plane loading but also other mechanical charges.

To recall, bolted joint usually consists of a bolt, two washers, two members and a nut (Figure I-85). When the compression is applied to a composite member and a metallic washer of smaller diameter (Figure I-95), the bearing damage can occur. It is possible to notice it visually due to the thickness change within the area of contact with the metallic part as a result of localised plastic deformation after the application of high pressure. In practice, it may happen during the preload of bolted joint when the tightening force exceeds the limit of bearing strength of the composite. Matrix out-of-plane compression is neglected in this work, whereas the onset of fibre damage may result in the irreversible changes. Proposed by CETIM, the criterion of maximum admissible compressive force consists in non-damaged reinforcement. This approach is adopted in this work. Hence, it is proposed to define the maximum compressive force by applying the compressive load to a composite specimen and a washer till the appearance of cracks in tows.

The composite specimens with diameter 36 mm (Figure II-9 d) and a metallic washer of 18 mm are used for the proposed experiment. The thickness of the composite is chosen 2 mm due to the limited maximum compressive capacity of a loading cell of the testing machine Instron 5584. Specimens are conditioned prior to the experiment at +90°C/RH 85% according to the developed protocol in section II.2. They are tested at the ambient temperature in an air-conditioned laboratory of GeM. The duration of the experiment is around 2 min.

The first experiment is conducted until the ultimate failure in order to obtain the maximum compressive force, which is nearly 51 kN. The splitting path is easily noticeable as indicated with the solid red line in Figure IV-15. The rest of the specimens are loaded from 35 kN till 50 kN with the step of 5 kN; they are then stopped to detect the appearance of cracks in fibres.

Following the experiments, the tested specimens are cut into two parts and analysed with SEM. One can see from Figure IV-15 that the significant deformation occurs for stresses close to the failure, for instance, at 40 kN and 45 kN. Although the fibres do not seem to be damaged in these cases, it is, however, highly probable in zones where the reinforcement is substantially closer to the material surface due to important deformation. The deformation is significantly reduced at the loading levels of 30 kN and 35 kN. Thus, it is decided to approve 35 kN as the preload force. This choice is also justified by the fact that if the preload is relatively small, its loss may be less detectable during the experimental characterisation. The selected loading is, however, expected to be dependent on the testing temperature and moisture content that results from the reduction of material mechanical properties according to presented results in Chapter II section II.4.



Figure IV-15: Compressive stress - displacement curve of the composite member of 2 mm thickness

IV.2.2.3. Bolt calibration

Bolts and nuts of trademark BUMAX® are made of high-tensile stainless steel with good resistance to high and low temperatures. The class of fasteners is 10.9. The geometry of bolts is introduced in Figure IV-16. All the bolts are instrumented with 2 uniaxial diametrically opposite strain gauges CEA-06-125UN-350 by PRESCAMEX to measure the shaft deformation during testing. The strain gauge configuration is the four-wire quarter bridge. The adhesive M-600 from Vishay is selected due to its resistance to harsh environmental conditions. The calibration is performed at CETIM Saint-Étienne.

The calibration of bolts prior to experimental testing is necessary in order to:

- Remove all the undesirable effects from gauge installation;
- Verify whether the bolt and the gauges perform linearly;
- Obtain an equation describing the mechanical behaviour of the bolt.

Hence, such a procedure provides the linear relation between applied tensile loading and deformation of a bolt at opposite sides. The coefficients of linear function are obtained (Eq. 66).

Prior to calibration, a clamped length is determined. Taking into consideration all the specimen configurations to test, the clamped length (the stretched length of a bolt) is 42 mm for all the bolts. It also must be respected during calibration. For the calibration, the head of a bolt is fixed in the upper grip, whereas the other end is locked into a special nut, installed in the lower grip (Figure IV-17). The working length is adjusted to be exactly 42 mm. The maximum level of applied tension (F_{Gi}) is 60 kN in order to avoid any bolt plasticisation due to the present drilling. The first traction is dedicated to the removal of all the undesirable issues resulting from the gauge installation. Then the following recorded traction is reproduced three times for the repeatability of results. According to a protocol of CETIM, the applied tensile load is maintained constant for 30 s at the

levels between 40 kN and 60 kN with the increment of 5 kN in order to register the bolt deformation (μDef). The linear relation between force and deformation enables computing parameters a and b (Figure IV-18). Consequently, each strain gauge describes the mechanical behaviour of the bolt. The calibration is performed at the ambient temperature and relative humidity. Bolts are not calibrated for torsion of the shank.

$$F_{Gi} = a \cdot \mu Def + b \tag{66}$$

Obtained parameters are then integrated into the software CATMAN Easy for each strain gauge. Hence, the preload can be applied till the desired level and its evolution can be directly seen while tightening. What is more, the changes in bolt elongation are assessed easily as presented in section IV.2.3.



Figure IV-18: Bolt calibration represented in Force (kN) – Microdeformation (μ def, 1000 μ def = 0.1%) diagrams for a) strain gauge 1 and b) strain gauge 2. Letter M stands for the loading order

IV.2.2.4. Bolt compliance

Calibration of bolts provides the coefficients for the linear relation between applied force and bolt deformation (Eq. 66). In order to compute the elongation of bolts during preloading instead of deformation, one must know the bolt compliance (Eq. 70 in section IV.2.3). Parameters, outlined in Table IV-5, are necessary for its evaluation. Their interpretation can be found in **[159,202]**.

Data of bolt					
Bolt diameter	d = 12 mm				
Thread pitch	$P = 1.75 \ mm$				
Elastic modulus	$E_b = 200 \ GPa$				
Geometrical bolt parameters					
Diameter of reduced shank	$d_{red} = 11 \ mm$				
Length of not-threaded shank under the bolt head	$l_{b \ head} = 5 \ mm$				
Length of reduced shank	$l_{red} = 15 \ mm$				
Length of non-threaded shank before threading	$l_{b \ thread} = 10 \ mm$				
Length of threaded shank except engaged	$l_{thr \ shank} = 12 \ mm$				
Diameter of engaged internal threads (Eq. 68)	$d_3 = 9.85 mm$				
Cross-section, associated with d_3 (Eq. 68)	$A_3 = 76.2 \ mm^2$				
Nominal bolt cross-section (Eq. 68)	$A_n = 113.1 \ mm^2$				

Table IV-5: Bolt parameters. Based on Figure IV-16

Bolt compliance is defined according to the equation [159,160]:

$$\delta_b^i = \frac{l_b^i}{E_b A_b^i} = \frac{1}{E_b} \left(\frac{0.4d}{A_n} + \sum_{i=1}^n \frac{l_{bi}}{A_{bi}} + \frac{L_p - \sum_{i=1}^n l_{bi}}{A_3} + \frac{0.5d}{A_3} + \frac{0.4d}{A_n} \right); \tag{67}$$

$$\begin{cases} d_3 = d - 1.2268 \cdot P = 9.85 \ mm; \\ A_3 = \frac{\pi \cdot d_3^2}{4} = 76.2 \ mm^2; \\ A_n = \frac{\pi \cdot d^2}{4} = 113.1 \ mm^2; \end{cases}$$
(68)

where the 1st expression in the parentheses stands for compliance of the bolt head, the 2nd is compliance of not threaded shank, 3^{rd} – compliance of threaded shank except engaged, 4^{th} – compliance of engaged internal threads of the bolt, 5^{th} – compliance of engaged external threads of the nut. According to the bolt geometry, illustrated in Figure IV-16, Eq. 67 is written as follows:

$$\delta_b = \frac{1}{E_b} \left(\frac{0.4d}{A_n} + \left[\frac{4l_{b\ head}}{\pi d^2} + \frac{4l_{red}}{\pi d_{red}^2} + \frac{4l_{b\ thread}}{\pi d^2} \right] + \frac{l_{thr\ shank}}{A_3} + \frac{0.5d_3}{A_3} + \frac{0.4d}{A_n} \right) \tag{69}$$

However, the theoretical formulation does not take into consideration the drilling in the shank. More precise estimation can be done with the software CETIM-Cobra, which is introduced in the literature review in section I.2.2.4. The bolt compliance is, therefore, determined with and without the drilling and outlined in Table IV-6. The difference between numerical and analytical calculations is 1.419 %. It may originate from the type of bolt head in the analytical expression. The coefficient 0.4 in Eq. 67 assumes that the head of the bolt is cylindrical whereas for the simulation in CETIM-Cobra the hexagonal bolt head is used. Apparently, the non-inclusion of drilling is not critical. Thus, the selected value of bolt compliance for the further computations is $3.030 \, 10^{-6} \, mm/N$ as estimated in CETIM-Cobra for hexagonal bolt head.

Bolt con	Variance	
Analytical computation	$\delta_b = 2.987 \ 10^{-6} \ mm/N$	-
CETIM-Cobra, without drilling	$\delta_b = 3.030 \ 10^{-6} \ mm/N$	1.419 %
CETIM-Cobra, with drilling	$\delta_b = 3.040 \ 10^{-6} \ mm/N$	1.743 %

Table IV-6: Bolt compliance

IV.2.3. Experimental results

This section outlines the results of the experimental campaign on bolted composite joints. Firstly, a trial test is carried out in order to reveal probable difficulties while performing the experiment and to eventually set it up. Subsequently, the test is run at two environmental conditions +23°C/RH 50% and +23°C/RH 85%. High testing temperatures are not applied due to the sensitivity of adhesive to the combination of temperature and humidity as graphically indicated at the end of the present section. As for the low temperatures, for instance, -40°C, such an experiment would not demonstrate the impact of the sensitivity of the composite bolted joint behaviour to humidity due to the high stiffness properties of the composite. Despite the varying moisture content, they remain similar at -40°C (section III.3.3) making the experiment irrelevant.

Results for unconditioned specimens

The term "unconditioned" signifies that the used specimens are neither dried nor aged prior to the experiment; thus, the initial material state is unknown. The target of the test, the results of which are demonstrated in Figure IV-19, is to detect the conformity of the results and whether the gauges respond correctly in order to finalise preparation for the experimental campaign.

The preload is applied with a torque wrench up to 35 kN of tensile force occurring in bolts. Resulting from the bolt calibration (section IV.2.2.3), the load is directly controlled during its application in the data acquisition software CATMAN Easy®. The duration of the experiment is 17 days. The setup was placed in a laboratory without regulated air-conditioning system; however, the average temperature was around 17-20°C.

All tested configurations are presented in the illustrations below, where the colour stands for a bolt number and the line type for a gauge 1 or 2. Generally saying, the results within one configuration are coherent, without important scattering. Some oscillations, visible in each curve, are suggested to originate from the sun rays during the daytime and consequent cooling during the nights. What is important to mention is that some strain gauges demonstrate relatively different behaviour from the rest. It is presumed to occur due to the uneven force distribution resulting from the next factors:

- Heterogeneous member structure, where some part is stiffer (tows) than the other (matrix);
- Presence of twisting moment while loading, for which bolts are not calibrated;
- Not precise position of one contact face in relation to another for 2 and more assembled members;
- Sliding of one member in relation to another;
- Diameter of clearance hole in comparison with bolt making the gap of 1 mm. It may lead to bending while loading;
- Presence of 3 contact faces in the entirely metallic joint;
- Manual tightening.



Figure IV-19: Decrease in preload of bolted joints with unconditioned composite material PA6 GF (a – d) and metal (e). G1 and G2 stand for a gauge number

Among the mentioned issues, occurring twisting moment, sliding and member structure cannot be eliminated or changed. The impact of the rest can be reduced by fixing the 3D-printed plastic inserts of 41 mm length on the bolt to compensate the gap. Made tiny notches on the side of composite specimens serve for their better positioning within the joint.

Uneven compressive stresses in the composite members are also confirmed by the different bolt deformation on its opposite sides. It is noticeable while loading; thus, the loading is stopped once one of the two gauges reaches the strain corresponding to 35 kN. What is more, there are visible zones with bearing damage on some specimens after unscrewing. Visually detected most damaged specimens are studied with a tomograph in GeM laboratory. The reconstructed specimen structure is presented in Figure IV-20. As one can notice, some tows underwent cracking at the loading level of 35 kN. It is assumed to occur mainly due to rigid/ductile zones in the composite material by the reason that the full RVE is not included in the compression zone. Although the other factors intensify the response of the strain gauges, taken actions, mentioned above, are intended to reduce the side effects in order to obtain the real mechanical response of bolted joints. What is more, after brief inspection, not all specimens undergo cracking. Hence, the previously defined loading of 35 kN is adopted for further experiments on the condition that the maximum attention and precision is given when applying preload.



Figure IV-20: Fibre breakage after applied preload: a) reconstructed specimen from Tomograph images; b) cracks in tows inside the specimen

Results for specimens at RH 50%/85% and +23°C

As mentioned in the previous sections, the aim of this experimental campaign is to reveal the changes in the behaviour of bolted composite joints subjected to humidity impact. The evolution of applied preload is presented in Figure IV-21 – Figure IV-24 through the bolt deformation and summarised in Table IV-7. Some more detailed graphs for +23°C/RH 50% can be found in Appendix G.

Analysing the graphs, one can notice two phases of joint loosening. The first is related to the spontaneous reduction in preload, which is observed to begin after tightening and to last for several hours, followed by the relative stabilisation of applied loading. The second phase can be characterised by a steady decrease in preload being proportional to the test duration. It is reached in 20-24 h after the beginning of the experiment. In the case of the entirely metallic joints (blue colour on the graphs), the stabilisation occurs more rapidly; therefore, the results seem to be in good accordance with **[150]**. Following the analysis and comparison of Figure IV-21 – Figure IV-24, one can conclude that:

- Application of second preload results in the lower loss of bolt deformation over the same period of time as compared to the initial loading;
- Preload relaxation is globally higher at RH 85% in comparison to RH 50% for all composite specimens;
- Loss of preload is smaller for thinner specimens of the same fibre-matrix ratio;
- Presence of the contact face (2 mm + 2 mm) causes higher decrease in preload in comparison with the single composite member (4 mm).

Although some undesirable effects occurred while loading, their impacts are expected to be of lower magnitude as for the test with unconditioned specimens, described in the previous section. Thus, sliding is decreased by reason of specimen precise positioning due to made notches. The benefit of tubular inserts is visible from the less different gauges response. However, the response of some gauges at RH 85% showed the constant increase of preload over time, which has no physical sense. Thus, one can presume that the adhesive layer is significantly impacted by high-level humidity. It may also occur due to not sufficient protective coating for strain gauges. The strain evaluation for these particular bolts is, eventually, not enhanced with the free bolt

used for compensation of environmental impact on the gauges. Hence, in order not to accumulate the errors, these bolts are not used for the computations of configurations "6 mm + 6 mm" and "4 mm + 4 mm", results of which are demonstrated in Figure IV-22 and Figure IV-24. It explains the unexpected position of these configurations with respect to the others since only 1 or 2 bolts are selected for the mean values.

In addition, brief testing of unloaded bolts at +80°C/RH 85% demonstrated the continuous increase of bolt deformation without any stabilisation over time. As a result, it is supposed that the plasticisation of adhesive is intensified under the coupled impact of high temperature and humidity. Hence, a proper choice of adhesive type is of vital importance for experiments at extreme conditions.



Apart from bolt deformation and preload evolution, bolt elongation can be estimated with the use of the bolt compliance, computed in section IV.2.2.4, and applied preload according to Eq. 70. This parameter is important to know for the design of bolted joints in order to ensure loading within the elastic limit after the preload application. According to Bickford **[202]**, designers traditionally use up to 0.2% of elongation to define the fastener yield stress, though this value may appear unreliable in some cases. Nevertheless, the application of minimum and maximum preload gives the range of bolt elongation resulting from tightening. Thus, the objective is to evaluate the bolt elongation for assembled composite members, which would contribute to more accurate design of bolted connexions via the software CETIM-Cobra.

$$L_b = \delta_b \cdot F_0^b \tag{70}$$

Englisher Sanaiman			Avera	ge of strain g	gauges	Average	e of bolt elong	ation
condition	config.	Load	E _{G start}	E _{G end}	Δε, %	L _{b start} , μm	L _{b end} , μm	ΔL _b , %
		1	1690.162	1291.410	23.552	84.373	67.127	20.427
	6 mm + 6 mm	1	± 176.503	±126.712	± 0.967	± 6.264	±4.976	± 0.752
	0 11111 + 0 11111	2	1917.318	1577.208	17.629	94.074	79.360	15.550
		2	±157.173	±84.793	±2.419	±5.917	± 3.039	±2.526
		1	1903.083	1406.438	25.750	92.396	71.297	22.633
	4 mm + 4 mm	1	± 381.380	± 232.802	±4.447	±15.175	± 10.000	±3.237
	7 11111 / 7 11111	2	1836.919	1580.230	13.966	89.621	78.621	12.266
		2	± 125.000	±105.316	± 0.121	±4.796	±4.073	±0.151
		1	1889.438	1543.177	18.355	92.749	77.719	16.207
+23°C	2 mm + 2 mm	1	±225.715	±189.916	± 0.997	± 7.907	± 6.658	± 0.932
RH 50%	2 11111 + 2 11111	2	2067.965	1797.156	12.905	100.213	88.457	11.605
		2	±194.372	±127.309	± 2.030	± 6.222	± 3.476	±2.019
		1	1798.475	1512.703	15.919	88.356	76.019	13.972
	1 mm	1	± 245.233	±213.168	± 0.415	± 9.340	±8.294	±0.439
	7 11111	2	1869.057	1736.823	7.138	91.447	85.689	6.329
		2	±192.454	± 204.272	±1.454	± 7.206	±7.692	±1.276
	metal	1	1867.926	1795.228	3.974	92.774	89.429	3.647
		1	±214.948	±231.869	±1.448	±8.432	±9.332	±1.690
		2	1846.257	1818.568	1.492	91.666	90.434	1.341
			± 276.741	± 270.929	± 0.098	±11.296	±11.094	±0.062
	6 mm + 6 mm	1	1870.506	1278.374	31.764	89.222	64.609	27.649
+23°C		1	± 256.632	±189.797	± 0.785	± 7.307	±5.964	± 0.759
RH 85%		_	1794.715	1457.562	18.786	89.728	74.695	16.754
		2						
			1913.927	1242.764	35.028	91.047	62.877	30.914
		1	± 642.088	±843.428	±16.647	± 26.530	± 34.769	±13.963
	4 mm + 4 mm		2014.713	1459.998	25.560	95.465	72.337	22.569
		2	±311.316	±25.513	±12.769	±13.865	±0.162	±11.415
			1976.907	1341.413	32.136	96.886	69.331	28.453
		1	± 22.303	± 68.450	± 3.481	± 1.120	± 3.626	± 3.441
	2 mm + 2 mm		2060.636	1466 239	27 697	100 244	74 664	24 793
±23°C		2	+263.743	+215.475	± 18709	+9.153	+11 237	± 16.982
TZJ C RH 85%			1027 236	1303 674	27.601	93 305	70.280	24.625
MII 05 70		1	+222.807	+159.074	± 27.001	+7.132	+5 279	± 2361
	4 mm		1020.260	1709.420	10.97	<u> </u>	-3.378	-2.301
		2	1920.360	1708.429	10.807	92.778	83.603	9.782
			±42/.//4	±293.084	±3.894	±22.06/	±16.539	±3.021
		1	1931.483	1868.323	3.151	95.623	92.825	2.817
	metal		± 378.823	± 337.061	±1.390	±18.869	±17.090	±1.180
		2	2102.973	2037.787	2.934	104.990	102.088	2.633
			± 306.396	± 273.470	±1.138	±14.060	±12.665	± 0.976

Table IV-7: Bolt deformation and elongation as a function of member thickness and environmental conditions

The computed decrease in elongation, which also shows the ratio of preload reduction, is illustrated in Figure IV-25 for the testing condition $+23^{\circ}$ C/RH 50% and in Figure IV-26 for $+23^{\circ}$ C/RH 85%. The results in micrometres are outlined in Table IV-7. The force F_0 is computed according to the equations obtained during

the bolt calibration with the use of data of strain gauges. It explains the similarity of results $\Delta \varepsilon$ and ΔL_b in Table IV-7. Some differences arise from the fact that some strain gauges are not loaded till 35 kN (more details in the previous section); thus, the average value of applied force is below 35 kN. In spite of the linear relations, the calibration was accomplished from 40 kN to 60 kN. Hence, the minor variations result from not absolutely precise coefficients of linear equations for studied loading levels.

35

+23°C/RH 85%

Load 1 and 2



load 1: 6 mm + 6 mm
 load 1: 4 mm + 4 mm
 load 1: 2 mm + 2 mm

load 1: 4 m

% load 2: 6 mm + 6 mm % load 2: 4 mm + 4 mm % load 2: 2 mm + 2 mm

load 2: 4 m

Figure IV-25: Comparison of reduction of bolt elongation after 1st and 2nd preloads at +23°C/RH 50%

Figure IV-26: Comparison of reduction of bolt elongation after 1st and 2nd preloads at +23°C/RH 85%

IV.2.4. Conclusions

This section outlines the methodology and the experimental campaign on the investigation of preload evolution in bolted joints with woven composite parts. Whatever the member material, the reduction of preload occurs in two stages: immediate and long-term loosening. The amount of loss can vary due to environmental conditions, level of initial preload, material of assembled parts, their geometry etc. Poor mechanical properties and high ductility of members lead to higher creep strain and lower bearing resistance that may also strongly deteriorate behaviour of bolted structures over time due to loosening. Thus, the aim of this experimental campaign was to study the preloaded bolted joint behaviour over time under different environmental conditions. This is particularly important for bolted composite joints due to the absence of a standard for such parts. What is more, the presently used standard for metallic materials does not account for humid impact.

The studied composite PA6 GF, experimentally and numerically investigated in Chapters II and III, manifests itself as highly ductile under certain environmental conditions. Consequently, preload estimation for this composite demonstrates the extreme cases of loosening under the given environment. Prior to launching an experimental campaign, some preparation work was carried out. It consisted of the evaluation of loading level, resulting from the low composite bearing strength and bolt calibration. The latter is of particular interest for the reason that it allows precise determination of bolt deformation and direct conversion from the deformation to the tensile force, occurring in the bolt. Thus, the loading level is controlled directly while tightening.

Obtained results, presented in the illustrations, demonstrate that the thick specimens and high humidity level result in the increased reduction of preload and bolt elongation. This was expected from the characterisation of member compliance, which turned to be higher for high temperature/humidity and thicker specimens. In addition, the presence of contact faces does not contribute to the conservation of preload as can be seen for the total member thickness of 4 mm. However, the application of the second loading leads to increased preload retention. As for the elongation of bolts after tightening, these values are important for an accurate composite joint design and are expected to be integrated into the software CETIM-Cobra.

CONCLUSIONS AND PERSPECTIVES

V.1. GENERAL CONCLUSIONS

The present research work is a doctoral thesis pursued in collaboration of Research Institute in Civil and Mechanical Engineering GeM and Technical Centre for Mechanical Industry CETIM. It focuses on the durability of prestressed bolted joints with clamped composite parts subjected to hygro-thermal conditions. The principal difference of bolted from other composite-metal connections is the possibility to delay the joint failure by applying preload. Its high level is necessary in order to avoid interface opening of tightened parts and to ensure fatigue endurance of fasteners. Therefore, the materials must sustain significant compressive stresses when in service. As demonstrated in the literature review, the out-of-plane creep deformation of composites with vulnerable properties, aggravated by environmental conditions, may affect the pre-tightening procedure of joints, which can lead to non-controlled consequences. The analysis of bolted joint behaviour, in particular, the evolution of preload over time, requires a considerable database of properties of clamped parts.

Prior to bolted joint characterisation, the experimental and numerical campaigns are proposed in order to identify the elastic mechanical properties of provided for the research work thermoplastic composite materials and neat matrix polyamide 6 (neat PPS is unavailable). Both composites, PA6 GF and PPS CF, are of balanced woven structure, but are targeted for applications in different domains due to the dissimilarity of chemical, physical and mechanical properties. Thus, working conditions vary from RH 0% to RH 85% and from -40°C to +80°C for PA6 GF, whereas the in-service temperatures of PPS CF are from -50°C to +120°C. Despite a great number of research articles on PA6 and PA6 GF, the absence of defined and sorted data at required environmental conditions leads to the necessity to perform experimental campaigns at different temperatures and for different RH levels. Similarly, not much attention of researches is given to PPS CF, presumably, due to its complex manufacturing process. Thus, the mechanical behaviour of three materials is to be estimated in order to a) create a database of properties under varying climatic conditions, b) to apply certain parameters for numerical simulations and c) to estimate their impact on the bolted joint. To attain these objectives, a rigorous experimental methodology is essential.

As presented in Chapter I, the hygroscopicity of PA6 GF and PA6 results in the significant changes of characteristics that are aggravated by the thermal impact and eventually form a coupled problem. Although a little quantity of moisture is absorbed by PPS CF composite, the elastic properties generally remain unaffected on the contrary to the influence of high temperatures; however, the elastoplastic behaviour may undergo some transformations. The change of properties is particularly visible in the vicinity of the glass transition temperatures that are around +60°C for PA6 and PA6 GF and around +90°C for PPS CF in dry states. However, the absorbed moisture causes a drop of T_g of PA6 and PA6 GF. Considering such changes of properties of the composites and the matrix, it is essential to ensure moisture content that would correspond to the surrounding atmosphere represented by RH. Hence, prior to any experimental testing, the conditioning protocol for desorption/absorption is developed and validated (Chapter II) in order to characterise materials in a reliable manner. The protocol is necessary due to the absence of details in the open access and, in addition, the information concerning the conditioning parameters is usually not provided by manufacturers. Thus, the proposed conditioning protocol allows controlling moisture content within specimens over time at a given RH. It is initially established for PA6 GF and verified with the use of PA6 specimens. Such choice is justified by the elevated moisture impact on this composite that makes it stand for a "reference". Consequently, the protocol is adjusted for the second composite material PPS CF. Its conditioning is essentially based on the fact that a rigorous study approach at some RH levels may demonstrate variations for the non-linear response, which is one of the widely investigated subjects in the research field. Hence, despite the interest in elastic properties, a conditioning methodology is likewise proposed for PPS CF. The temperature, used for specimen conditioning, is approved by the tensile shear tests that do not reveal significant material changes; nonetheless, the prolonged

exposure may eventually result in chemical and physical degradation that is not examined in the work. Therefore, the temperature +90°C is selected for desorption and absorption of all three materials, which greatly accelerates the kinetics in comparison to other evaluated temperatures. The duration of conditioning, outlined in Chapter II, may vary if material thickness changes. Thus, it is important to consider the geometry of specimens before the protocol application. Owing to the conditioning and appropriate moisture content, the glass transition temperature, driven by matrix, is successfully examined for PA6 GF at 0%, 50% and 85% of RH. The results confirm that the composite state tends to change from a rigid at RH 0% ($T_q \approx +57^{\circ}$ C) to a viscous at RH 85% $(T_g \approx -1^{\circ}C)$. Under the standard ambient conditions (RH 50% and +23°C), the glass transition temperature is around +23°C. According to the literature review for the composite PPS CF, its T_g does not have a tendency to change due to moisture uptake, thus, it is not analysed in this work. Nevertheless, known T_q enables better comprehension and analysis of mechanical behaviour of the composites and the matrix, studied during the experimental campaign for the in-plane characterisation. The conditioning protocol is applied to all the specimens. The elastic, ultimate and damage parameters of each material are evaluated under different testing conditions; however, the latter are not presented due to the main focus of the present work on the elastic behaviour. The computed properties reveal the coupled environmental impact on the durability of materials. The increase of temperature and moisture content leads to a drop of shear moduli, shear strength and yield shear stress of PA6 GF. The shear strain seems to increase till a threshold, thus emphasising ductile properties of the composite above its T_q . The longitudinal and transversal properties do not undergo significant changes under the thermal and humid impact. The same tendency to increasing strains, decreasing stresses and moduli is observed for PA6 under hygro-thermal conditions. As for the composite PPS CF, the water impact on its shear properties is practically non-existent in the elastic region, but the working temperature leads to a similar reduction of properties as those of PA6 GF. Hence, the conditioning protocol for PPS CF is not of absolute necessity for the evaluation of elastic parameters, but it is still suggested for the investigation of elastoplastic behaviour. The longitudinal and transversal moduli seem to experience minor changes only at +120°C. However, it can be related to the natural dispersion of results during experiments. The monitored mass of specimens illustrates a material capacity to a rapid moisture loss or gain during a short period and remains in accordance with the results of desiccation/absorption protocol. Globally saying, the obtained results are coherent with the literature. Consequently, the studied environmental impact on the mechanical behaviour of materials allows the construction of a reliable experimental database. Furthermore, the result of matrix PA6 and the composite PA6 GF are used for the development and validation of numerical modelling procedure in Chapter III and analysis of bolted joint behaviour over time in Chapter IV. The properties of PPS CF are not used hereafter due to the unavailability of neat PPS behaviour at the same environmental conditions.

The experimental characterisation in Chapter II provides a wide range of in-plane properties, but the out-ofplane parameters are difficult to assess experimentally. Besides, the through-thickness behaviour represents the main interest for the pre-stressed bolted joints. Thus, a numerical approach is proposed for the identification of necessary properties. It is based on the double-scale homogenisation technique with the application of the finite element method at a scale of a tow and a composite. The properties of elementary constituents of composite material are employed to reproduce the Representative Volume Element (RVE) of the composite PA6 GF, where the experimentally computed properties of the neat PA6 are adopted for the matrix representation. The full RVE geometry is reconstructed with the use of images from Scanning Electron Microscope at different magnifications. They enable to calculate tow dimensions and fibre content as well as the positioning of tows through the thickness. Although an RVE model for each composite material is generated, the only RVE of PA6 GF serves for the homogenisation. The elastic parameters of tows are computed at the micro-scale (fibre and matrix) on the periodic cubic structure. They are consequently integrated into the full-scale RVE. Such simulation provides the whole range of numerical results that confirm the hygro-thermal impact on the out-ofplane behaviour of the composite material. The in-plane numerical outcome is globally in agreement with the experimental, except at negative temperatures. As mentioned, it can be related to some strong assumptions for the numerical simulations, where the uniform moisture distribution and the choice of periodic tow RVE seem to underrate the material modulus at negative temperatures. Nevertheless, the computed out-of-plane properties are in accordance with the experimental in-plane behaviour. Hence, the modulus E_{33} is employed for the evaluation of bolted joint behaviour in Chapter IV for the reason that its performance is highly dependent on the out-of-plane rigidity of clamped parts.

It can be suggested that the decreasing out-of-plane modulus of the composite may lead to rapid loss of preload due to the increasing part compliance. The decrease of pre-stress can be intensified by the environmental conditions. Their coupled impact may lead to the overall degradation of mechanical performance of bolted joints. In order to provide the true values of composite part compliance at given environmental conditions, the analytical and numerical approaches are used and compared. Numerical results exceed those, calculated analytically, especially for considerable member thickness. It is suggested to be related to the heterogeneous orthotropic structure, where the deformation cone practically vanishes. Besides, the characterisation of preload demonstrates the high pretightening loss in joint with thick members. Although the preload loosening is dependent on its initial level, geometry of clamped members, their number, low bearing resistance and high material ductility are the principal arguments. The environmental impact results in higher preload reduction. Therefore, the application of second loading tends to ameliorate the preload retention for the same period. To conclude, the change in mechanical properties of clamped parts directly affects the durability of preloaded bolted joints. Therefore, additional preload characterisation for composite members remains necessary.

V.2. **PERSPECTIVES**

A wide range of subjects, related to the thermoplastic composite materials and their bolted joints, are investigated in the presented PhD thesis. Nevertheless, there is some work which remains to be accomplished in order to enhance the potential of the research. It is proposed to be performed on two fronts: experimental and numerical. Besides, the perspectives are classified as short-term and long-term.

The short-term perspectives are suggested in order to:

- Expand the conditioning protocol so to take into consideration specimen thickness;
- Estimate more precisely the out-of-plane hygroscopic coefficient by applying other methods;
- Extend the range of obtained results for PPS CF by examining not studied environmental conditions, for instance, negative temperatures;
- Perform the experimental campaign on the neat matrix PPS with the subsequent possibility to apply the double-scale homogenisation to the composite material PPS CF;
- Optimise the computed homogenised values by their validation on the several RVE types of a tow;
- Define the quasi-static velocity by considering low strain rate loading for the mechanical response of 45°-fibre oriented specimens and the matrix. Besides, the established experimental database can be supplemented by additional results;
- Revise the data acquisition and processing methods in order to ameliorate the precision of results. It concerns the Data Image Correlation method, which is not entirely appropriate for small strains occurring in 0° and 90°-fibre oriented specimens. As for the large strain of 45°-fibre oriented specimens, the DIC method and extensometers are preferred. Regarding the data processing, a more accurate definition of the linear zone is necessary, particularly for the ductile materials. The use of the method of least squares enables the selection either of maximum R^2 values or of the fixed value (for instance, $R^2 = 0.995$) which results in different properties. This approach should be re-examined in order to provide a clear, consistent and accurate methodology for the evaluation of elastic mechanical properties.

The long-term perspectives concern:

- More detailed experimental characterisation of bolted composite joints regarding the impact of environmental conditions. Besides, the effects of higher/lower and cyclic loading on tightened bolted joints can be further explored. The analyses of the plastic deformation in composite parts caused by the metallic washer could serve to evaluate the limitations of preload to absolutely avoid material damage if required. In addition, the variation of part and washer geometries might widen the database of CETIM-Cobra, used for the joint dimensioning;
- Cyclic environmental impact. The cyclic desorption/absorption reveals some changes in the mass of the composite specimens as briefly presented in this work. It may be interesting to study this feature in more detail for the reason that it may result from chemical or physical changes and lead to aggravated mechanical properties for long-term applications;
- Complete identification of mechanical behaviour of the composite materials as a function of environmental conditions. It involves the non-identified irreversible deformations and damage parameters briefly demonstrated in Appendix D. Thus, the constitutive laws of the composite materials can be established with the relation to environmental conditions;

- Homogenisation with more accurate moisture distribution within the composite material and the tow. Besides, the contact in fibre-matrix and tow-matrix zones, presumed as perfect, should be revised and described in a more precise manner;
- Extension of calculations of bolted joints with clamped composite parts. The numerical modelling of composite part compliance necessitates the introduction of the interface surface. It would expand the computations to the multi-material bolted joints.

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APPENDICES

A.Protocol validation for PA6 GF



Figure VII-1: Moisture desorption as a function of the square root of time



Figure VII-2: Moisture absorption at RH 85% as a function of the square root of time. Circular specimens


Figure VII-3: Moisture absorption at RH 50% as a function of the square root of time. Circular specimens



Figure VII-4: Moisture absorption at RH 50% as a function of the square root of time divided by specimen thickness. Circular specimens

Neat matrix PA6



B. Graphs for hygroscopic swelling

Figure VII-5: Out-of-plane hygroscopic deformation of matrix specimen of type 2 at RH 50% and RH 85% as a function of moisture uptake



Figure VII-6: Out-of-plane hygroscopic deformation of matrix specimen of type 3 at RH 50% and RH 85% as a function of moisture uptake



Glass-fibre reinforced polyamide 6

Figure VII-7: Out-of-plane deformation of circular specimens PA6 GF as a function of C_{glob} when subjected to RH 85%



Figure VII-8: Out-of-plane deformation of circular specimens PA6 GF over time when subjected to RH 85%



Figure VII-9: Out-of-plane hygroscopic deformation of circular specimens PA6 GF of 2 mm thickness as a function of C_{glob} when subjected to RH 85%



Figure VII-10: Out-of-plane hygroscopic deformation of circular specimens PA6 GF of 4 mm thickness as a function of C_{glob} when subjected to RH 85%



Figure VII-11: Out-of-plane hygroscopic deformation of circular specimens PA6 GF of 6 mm thickness as a function of C_{glob} when subjected to RH 85%



C. Dynamic mechanical analysis of PA6 GF

Figure VII-12: DMA of desiccated specimen 1 of PA6 GF. Corresponds to RH 0%



Figure VII-13: DMA of desiccated specimen 2 of PA6 GF. Corresponds to RH 0%



Figure VII-14: DMA of desiccated specimen 1 of PA6 GF. Corresponds to RH 50%



Figure VII-15: DMA of desiccated specimen 2 of PA6 GF. Corresponds to RH 50%



Figure VII-16: DMA of desiccated specimen 1 of PA6 GF. Corresponds to RH 85%

D.In-plane experimental campaign



Glass-fibre reinforced polyamide 6





Figure VII-18: Loss in mass of reference specimens of PA6 GF 45°



Figure VII-19: Comparison of shear modulus of PA6 GF tested at 2 mm/min and 0.5 mm/min



Figure VII-20: Comparison of shear stress at yield of PA6 GF tested at 2 mm/min and 0.5 mm/min

Damage and plasticity characterisation

For the cyclic tests (Figure VII-21), the total material deformation is composed of elastic and irreversible strains as presented in Eq. 71 for 45°-fibre oriented composite material. One can characterise damaged material state by computing damage parameters d_{12}^i for each cycle taking the slope between the minimum and maximum of the cycle. The damage-associated thermodynamical force Y_{12}^i characterises the propagation of damage and is estimated according to Eq. 73. The corresponding pair of data (d_{12}^i, Y_{12}^i) , coming from the different hysteresis loops, allow to form the curve $Y_{12} = f(d)$. An approximation, which can be linear, logarithmic..., is done in order to determine the form for damage evolution law.

Concerning the evolution of the irreversible strain, pair of data (p^i, R^i) can be computed using the irreversible strain and its associated damage as well as the yield function. The cumulative plastic strain can be estimated as stated in Eq. 74. The yield function is given by Eq. 75. This pair of data allows forming the curve R = f(p) for which a power law is often used as an approximation.

More details about data manipulation can be found in [203,204].

$$\gamma_T = \gamma_{12}^e + \gamma_{12}^p \tag{71}$$

$$d_{12}^{i} = 1 - \frac{G_{12}^{i}}{G_{12}^{0}} \tag{72}$$

$$Y_{12}^{i} = \sqrt{\frac{1}{2}} G_{12}^{0} (\gamma_{12}^{e})_{i}^{2}$$
⁽⁷³⁾

$$p = \int_{0}^{\gamma_{12}} (1 - d_{12}) \, d\gamma_{12}^p \tag{74}$$

$$R = \int_{0}^{\gamma_{12}} (1 - d_{12}) \, d\gamma_{12}^p \tag{75}$$



Figure VII-21: Theoretical shear stress - shear strain curve resulting from cyclic loading



Figure VII-22: Stress-strain response of PA6 GF at +23°C during cyclic quasi-static loading



Figure VII-23: Stress-strain response of PA6 GF at RH 0% during cyclic quasi-static loading



Figure VII-24: Stress-strain response of PA6 GF at RH 50% during cyclic quasi-static loading







Figure VII-26: Tensile-ruptured specimens PA6 GF of 0° at +80°C and RH 50%





Figure VII-28: Tensile-ruptured specimens PA6 GF of 45° at - $40^{\circ}\mathrm{C}$ and RH 0%



Figure VII-29: Tensile-ruptured specimens PA6 GF of 45° at $+80^{\circ}\mathrm{C}$ and RH 85%



Figure VII-30: Tensile-ruptured specimens PA6 GF of 45° at +23°C and RH 50%

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Carbon-fibre reinforced polyphenylene sulphide





Figure VII-32: Stress-strain response of PPS CF at RH 85% during cyclic quasi-static loading



Figure VII-33: Tensile-ruptured specimens PPS CF of 0° at $+23^{\circ}\text{C}$ and RH 0%

Figure VII-34: Tensile-ruptured specimens PPS CF of 90° at $+120^{\circ}C$ and RH 0%



Figure VII-35: Tensile-ruptured specimens of 45° at +105°C and $$\rm RH\ 85\%$$



Figure VII-36: Tensile-ruptured specimens of 45° at +90°C and RH 85%



Figure VII-37: Loss in mass of reference specimens of PPS CF 45°



Figure VII-38: Change in mass of reference specimens of PPS CF at RH 0% oriented at 0° and 90° $\,$



Neat matrix PA6





Figure VII-40: Comparison of tensile stress at yield of PA6 tested at 2 mm/min and 0.5 mm/min



Figure VII-41: Loss in mass of reference specimens of PA6

E. Comparison of DIC by VIC 2D, TEMA and gauges

R ² _{max}			VIC 2D			TEMA
	Specimen №	size 1	size 2	size 3	Gauges	avg. of 3 num.
		$16 imes 16 \ mm^2$	$10 imes 10 \ mm^2$	$25 imes 25 \ mm^2$		gauges
<i>G</i> ₁₂ , MPa	1	1697.881	1697.881	1702.716	2008.362	1739.598
	2	1429.923	1425.605	1386.604	1647.834	1278.329
	3	1929.220	1931.740	1887.415	1998.196	1923.062
σ_{12} , MPa	1	3.351	3.351	3.351	3.350	3.351
	2	3.053	3.053	3.053	3.052	3.054
	3	2.484	2.484	2.484	2.484	2.485

120 120 specimen 1 specimen 2 **True shear stress 012 true, MPa** 00 00 00 00 True shear stress 012 true, MPa 100 80 60 VIC 2D size 1 VIC 2D size 1 40 VIC 2D size 2 VIC 2D size 2 VIC 2D size 3 VIC 2D size 3 20 tema tema gauges gauges 0 0 0 10 20 30 40 50 60 0 10 20 30 40 50 60 True shear strain γ12 true, % True shear strain γ12 true, % b) a) 120 specimen 3 **True shear stress 012 true, MPa** -VIC 2D size 1 VIC 2D size 2 VIC 2D size 3 tema gauges 0 0 10 20 30 40 50 60 True shear strain γ12 true, % c)

Table VII-1: True mechanical values as a function of data processing methods for tensile tests of unconditioned PA6 GF

Figure VII-42: True stress-strain curves as a function of data processing methods of unconditioned PA6 GF



F. Compliance of composite parts

Figure VII-43: Composite compliance as a function of environmental conditions and cone angle φ for 6 mm thickness



Figure VII-44: Composite compliance as a function of elastic modulus and cone angle φ for 6 mm thickness

G. Loss of pretension in bolted joints at+23°C/RH 50%



Figure VII-45: Decrease in preload of bolted joints with 6 mm + 6 mm PA6 GF at +23°/RH 50%. Load 1



Figure VII-47: Decrease in preload of bolted joints with 4 mm + 4 mm PA6 GF at +23°/RH 50%. Load 1



Figure VII-49: Decrease in preload of bolted joints with 2 mm + 2 mm PA6 GF at +23°/RH 50%. Load 1



Figure VII-46: Decrease in preload of bolted joints with 6 mm + 6 mm PA6 GF at +23°/RH 50%. Load 2



Figure VII-48: Decrease in preload of bolted joints with 4 mm + 4 mm PA6 GF at +23°/RH 50%. Load 2



Figure VII-50: Decrease in preload of bolted joints with 2 mm + 2 mm PA6 GF at +23°/RH 50%. Load 2



Figure VII-51: Decrease in preload of bolted joints with 4 mm PA6 GF at +23°/RH 50%. Load 1



Figure VII-53: Decrease in preload of metallic bolted joints at +23°/RH 50%. Load 1



Figure VII-52: Decrease in preload of bolted joints with 4 mm PA6 GF at +23°/RH 50%. Load 2



Figure VII-54: Decrease in preload of metallic bolted joints at $+23^{\circ}/RH$ 50%. Load 2

UNIVERSITE BRETAGNE SCIENCES LOIRE POUR L'INGENIEUR



Titre : Durabilité des performances mécaniques des assemblages boulonnés précontraints en composites dans un environnement hygro-thermo-mécanique

Mots clés : Composite thermoplastique, température, humidité, vieillissement, assemblage boulonné, précharge

Résumé : Ces travaux, menés en collaboration avec le CETIM, portent sur la performance mécanique des assemblages boulonnés précontraints en matériaux composites pour les secteurs automobile et aéronautique. En raison de l'application contrôlée de précontrainte, les assemblages boulonnés occupent une partie importante dans l'industrie. Les conditions environnementales de service ont tendance à varier au cours du temps, ayant un effet sur les composites thermoplastiques sensibles à la température et à l'humidité. Une estimation des propriétés élastiques hors plan des composites est donc essentielle pour dimensionnement précis des assemblages un boulonnés. L'objectif de la thèse est de prendre en considération et d'analyser l'impact environnemental sur les matériaux composites thermoplastiques afin d'améliorer un modèle analytique d'assemblages boulonnés.

Des protocoles de conditionnement sont proposés afin d'évaluer d'une manière précise l'état des matériaux à plusieurs niveaux d'Humidité Relative. L'effet du vieillissement humide est étudié à travers la caractérisation mécanique d'une résine pure et de deux matériaux composites tissés. Les simulations numériques fournissent les propriétés hors plan en lien avec les effets environnementaux permettant l'estimation de la souplesse des matériaux. Des essais mécaniques sur les assemblages boulonnés en composites sont proposés afin de déterminer la perte de précontrainte au cours du temps et de relier les propriétés mécaniques des composites à la durabilité des assemblages.

Title : Durability of mechanical performance of prestressed bolted composite joints in a hygro-thermomechanical environment

Keywords : Thermoplastic composite, temperature, humidity, ageing, bolted joint, preload

Abstract : The present work, pursued in collaboration with CETIM, is focused on the preloaded mechanical performance bolted of composite joints for automotive and aeronautical fields. Owing to the controlled preload application, bolted joints occupy a significant segment in the industry. Working environmental conditions tend to vary over time, affecting sensitive to temperature and humidity thermoplastic composites. An estimation of out-of-plane elastic properties of composites is, therefore, essential for an accurate dimensioning of bolted joints. The objective of the thesis is to take into consideration and analyse the environmental impact on woven thermoplastic composite materials in order to ameliorate an analytical model for bolted composite joints.

Conditioning protocols are proposed for an accurate evaluation of material state at several Relative Humidity levels. Effect of humid ageing investigated through the performed mechanical characterisation of a neat thermoplastic matrix and woven two composite materials. Numerical simulations provide the out-of-plane environmentrelated properties enabling the material compliance estimation. Mechanical testing of bolted composite joints is proposed to determine the loss of preload over time and to relate composite mechanical properties to the durability of joints.