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INGENIEROS DE CAMINOS, CANALES Y PUERTOS

**Prediction of mechanical properties of
unidirectional FRP plies at different
environmental conditions by means of
computational micromechanics**

TESIS DOCTORAL

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Camino, Canales y Puertos

Universidad Politécnica de Madrid

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*A Ruth,
a mi hermano Alfonso
y a mis padres, José Antonio y María del Carmen.*

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Resumen

Actualmente, los materiales compuestos de matriz polimérica reforzados con fibras cerámicas son extensamente usados en aplicaciones que requieren excelentes propiedades específicas. Hasta ahora, varios modelos de fallo, con base física o fenomenológica, han sido desarrollados para predecir el comportamiento mecánico de láminas unidireccionales de dichos materiales. No obstante, estos modelos requieren un importante número de propiedades que han de ser obtenidas mediante largas y costosas campañas experimentales. Este es el caso de las certificaciones aeronáuticas, donde el proceso completo puede extenderse durante más de un año, debido especialmente a los ensayos de materiales envejecidos bajo diferentes condiciones ambientales. Por este motivo, la industria contempla cada vez más la realización de ensayos virtuales buscando una reducción de dichas campañas experimentales, actualmente imprescindibles en la caracterización de materiales compuestos a diferentes escalas que abarcan desde pequeñas probetas a componentes estructurales.

En esta tesis, se ha desarrollado una estrategia que combina experimentos y modelos numéricos dentro del marco de la micromecánica de materiales compuestos. Dicha estrategia ha permitido la predicción de las propiedades mecánicas de una lámina unidireccional de material compuesto cuando éste está en servicio bajo diferentes condiciones ambientales. Para ello, las principales propiedades de los constituyentes han sido medidas mediante una serie de ensayos micromecánicos desarrollados in-situ, es decir, en la propia sección transversal del material compuesto. En el caso de la matriz polimérica, la indentación instrumentada ha sido combinada con la compresión de micropilares para determinar el comportamiento elasto-plástico de la resina evitando cualquier dependencia de ensayos macroscópicos. Por otro lado, la resistencia de la intercara fibra/matriz ha sido medida mediante ensayos de push-in.

Respecto al trabajo computacional, una serie de microestructuras estadísticamente

equivalentes a la lámina real han sido virtualmente generadas usando la información obtenida del análisis micrográfico de la sección transversal del compuesto. La información resultante se ha empleado para generar un modelo tridimensional de elementos finitos de un volumen representativo de la lámina del material compuesto. El mecanismo de decohesión de la interfaz fibra/matriz ha sido simulado mediante un modelo cohesivo acoplado con fricción, mientras que un modelo elasto-plástico dependiente de la presión que incluye daño por tensión y compresión ha sido empleado para capturar el comportamiento no lineal de la matriz polimérica. La respuesta mecánica de ambos constituyentes ha sido calibrada usando los resultados de los ensayos micromecánicos descritos anteriormente. Este tipo de estrategia proporciona información relevante acerca del papel que juegan los diferentes constituyentes en los diferentes modos de fallo de la lámina. Además, proporciona las curvas tensión-deformación (sin necesidad de usar parámetros de ajuste) y reproduce los mecanismos de fallo observado experimentalmente para casos de carga uniaxial y biaxial.

El trabajo desarrollado en esta tesis sienta las bases necesarias para desarrollar una estrategia multiescala que permitirá, entre otras cosas, el desarrollo de ensayo virtuales de partes específicas de componentes, el estudio de la combinación de diferentes fibras y matrices y de la importancia de los defectos derivados de la fabricación. El resultado, será una reducción de costes y de tiempo, favoreciendo el uso de nuevos y mejores materiales adaptados a diferentes aplicaciones ingenieriles.

Abstract

Fiber Reinforced Polymers (FRP's) are nowadays extensively used in applications where very good mechanical properties are required in combination with weight savings. Up to date, various phenomenological as well as physically-based failure models have been proposed for unidirectional (UD) composites. Unfortunately, their input parameters have to be obtained through costly and time-consuming experimental campaigns. This is the case of certification in the aeronautical industry, where the whole process for each material can last well over a year due to the required tests for different environmental and ageing conditions. For this reason, virtual-testing of composites is getting more and more attention from the industry as it promises to reduce the experimental programs devoted to the characterization of composites at several scales, from coupons to components.

In this thesis, a coupled experimental-computational strategy based on a micromechanics framework was developed to predict the mechanical properties of a composite ply (stiffness and strength) under different operating environmental conditions. To this end, the main properties of material constituents are determined by means of *in-situ* (in the sense that they can be performed on the actual composite material) micromechanical measurements. Regarding the polymer matrix, two micromechanical techniques, instrumented indentation and micropillar compression, are combined to determine with high accuracy the polymer elasto-plastic behavior avoiding any type of macroscopic test dependency. On the other hand, fiber/matrix interface strength is measured by means of push-in tests.

Regarding the computational work, a set of virtual microstructures, statistically equivalent to the real ply, are generated using the information extracted from micrograph analysis of the composite cross-section. The resulting information is used to generate a 3D FE model of a Representative Volume Element (RVE) of the

composite ply. The interface decohesion is simulated using cohesive-frictional interactions, whereas a pressure dependent, elasto-plastic model that includes tensile and compression damage is employed to capture the matrix nonlinear behavior. The mechanical response of both constituents is calibrated using the properties measured by micromechanical testing above mentioned. This type of approach provides information regarding the importance of each constituent in the different failure modes and is able to reproduce the stress-strain curves (without fitting parameters), as well as the deformation and fracture mechanisms experimentally observed for uniaxial and biaxial loading cases.

The work developed in this thesis constitutes the building block of a bottom-up multiscale approach setting the basis for in-situ virtual testing of specific regions of the ply, fast characterization of different combinations of matrices and fibers and study of micro-defects resulting from deficient curing conditions, among other important industrial requirements. This leads to the reduction of time and costs involved in new material designs and provides fast screening capabilities that improve material selection in engineering applications.

"If I have seen further, it is by standing on the shoulders of giants".

Isaac Newton (1676 AD)

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Introduction and objectives

1

1.1 Composite materials and industry requirements

Composite materials, often shortened to composites, are most often defined as complex materials made from two or more constituents with significantly different physical or chemical properties. Smart combinations of different constituents can provide a desired response of the composite making it perfect for a particular given application. For this reason man-made composites have a long history, Egyptians already built houses using wattle and daub and ancient Mesopotamians realized that gluing wood at different angles the resulting plywood exhibited better properties than the ordinary ones. Thanks to this tailoring capability, different types of composites can be found nowadays in aerospace, automobile, construction, sports and many others technological industries.

Regarding lightweight structural applications, such as spacecraft and aircraft structures, fiber reinforced polymers (FRPs) are the most extensively used among all the high performance composites. FRPs are made by a polymer matrix reinforced with stiffer and stronger continuous fibers (carbon, glass, aramid, etc) that can be weaved, knitted or braided in order to create a textile sheet that will ease the manufacturing process. However, for high-end applications requiring high strength and stiffness composite laminates made by stacking several unidirectional (UD) plies are still the reference configuration. Each ply has millions of fibers running in a single direction, and the resulting material is highly anisotropic. The fibers are the load-bearing phase, while the matrix is mainly used to maintain the fibers oriented according to the design direction. The properties of the final laminate will depend on the stacking sequence of the individual plies. Therefore, specific laminates can be tailor in order to fulfill the desired properties.

1.1. Composite materials and industry requirements

The polymer matrix in FRPs is usually a thermoplastic or thermoset resin and their mechanical properties depend on the chemico-physical characteristics and the manufacturing processes. Thermoset resins are made from molecules with relatively small molecular weight called prepolymers. Since the length of these molecules is short, the material is initially a liquid of low viscosity at room temperature and, therefore, they are specially suited for processing of FRPs. The liquid resin is injected or infused into the fibers preforms and easily flows through the reinforcement, obtaining good impregnation of the fibers with low porosity. Once the fiber reinforcement is completely saturated, the resin is cured to make it solid. In order to cure the thermoset, the prepolymers must be tied together with other molecules. This process, known as cross-linking, occurs under the appropriate conditions of temperature and pressure. Thus, the resin is finally transformed into a solid, by the 3D chaining network created between the prepolymers. Since the cross-linking is a nonreversible process, the shape of the created component cannot be changed after curing. The most common polymeric matrices for high performance and aircraft grade composites are the epoxy resins. They have been extensively used due to their excellent properties such as good adhesion, good corrosion resistance, low shrinkage and processing versatility. On the other hand, epoxy resins present some important drawbacks, such as their cost and higher viscosity than other thermosetting resins, which results in more intricate and costly manufacturing processes. Therefore, the epoxy is only employed in high added-value applications such as aircraft, sports, etc.

A promising alternative to epoxy resins are the thermoplastics. In this type of resins the polymer chains are joined by intermolecular or van der Waals forces forming linear or branched structures. These intermolecular bonds weaken rapidly when increasing the temperature, yielding a viscous liquid. At this point the polymer can be formed and consolidated in minutes, unlike thermosetting resins which need large and expensive curing processes. In addition, thermoplastics can melt and be reformed several times unlike thermosetting resins, which do not melt but decompose and do not reform upon cooling. Therefore, thermoplastics are very suitable for processing techniques such as extrusion, compression and injection molding and calendering. Moreover, thermoplastics are known for their weldability, toughness, long storage life, recyclability and chemical resistance among other goodnesses. However, thermoplastic resins usually require high processing temperatures ($T > 350^{\circ}\text{C}$), leading to large thermal residual stresses during cooling due to the mismatch between the thermal expansion coefficients of matrix and reinforcements. In addition, above

its glass transition temperature, T_g , and below its melting point, T_m , the physical properties of a thermoplastic change drastically without an associated phase change. Therefore, the processing temperature range is quite narrow ($\Delta T \approx 30^\circ\text{C}$) and needs to be controlled carefully in the whole sample during the forming process, which is a formidable task. Finally, the cooling process affects the formation of intermolecular interactions and hence, controls the degree of crystallization, which will define the mechanical, optical and chemical response of the polymer [Parlevliet et al. \(2006\)](#). Therefore, cooling needs to be carefully controlled as well.

Although both types of polymers are suited for producing high-end FRPs, for structural applications where outstanding stiffness and strength are required, epoxy resins are still the best choice due to their superior strength, resistance to high temperature, dimensional stability and ease of processing compare to thermoplastics.

Regarding the type of reinforcements, long continuous carbon or glass fibers are the most common option for structural applications. In particular, carbon fibers offer the best specific properties (Figure [1.1](#)).

As an example of the growing usage of FRPs in modern aircraft industry, the new Boeing 787 Dreamliner and the Airbus A350 XWB are approximately 80% composite, by volume. In particular, their wings and fuselage are primarily made of carbon-fiber-reinforced polymers (CFRPs). In view of this, predicting mechanical properties and failure mechanisms of CFRPs becomes mandatory in order to prove the safety of structures upon whose integrity human lives depend. However the magnitude of this task is immense in terms of cost, time and human resources. The standard strategy to tackle the problem involves a material characterization through the different levels of the so called testing pyramid (Figure [1.2](#)). this building block approach requires massive testing campaigns starting at the ply coupon level. As we climb the pyramid the complexity and cost rapidly increase and, hence, the number of tests should be limited. Although this approach has proved its robustness in the last decades, the tardy certification process of new materials makes the aeronautical industry extremely conservative limiting the usage of innovative materials that can improve the performance of the aircraft. As an example, the prototype A350 first flew on 14 June 2013, but the A350 entered entered service with Qatar Airways on 15 January 2015, almost two years later.

Computational models can be used to ease the decision-making at the different levels of the testing pyramid. However the highly non-linear response of composites, once damage or fracture are initiated, hinders the prediction of the final strength

1.1. Composite materials and industry requirements

of the studied material. Extending this process for different loading conditions and environments requires the proper characterization of failure mechanisms at different scales acting from the atomic level up to the structure itself. Taking advantage of the hierarchical structure of composite materials the so called multiscale simulation techniques are growing faster during the last years. This multiscale simulations are aimed to reduce the number of tests required to obtain reliable predictions at the different pyramid levels using computational models.

(a)

(b)

Figure 1.1: Ashby charts comparing mechanical properties of composites with other materials.(a) Specific stiffness versus specific strength and (b) strength versus fracture toughness. Carbon and glass fiber-reinforced polymers (CFRP and GFRP, respectively) are highlighted. Adapted from [LLorca et al. \(2013\)](#)

1.2 Multiscale modeling: towards virtual testing

So far, the standard way to alleviate the characterization problem is the so called *global-local* approach, where material testing begins studying the composite structure and identifying the weakest parts where the damage is most likely to appear. Then, these regions are subjected to further refined analysis using nonlinear constitutive models to predict the material response up to the final fracture. However, this strategy mainly relies on phenomenological models where the constitutive relations at certain levels will depend directly on calibration tests performed on upper scales, where complexity and costs are higher. Due to the phenomenological nature of these models, input parameters have to be obtained on each material and the results obtained for a given unidirectional FRP ply can not be extrapolated to a material with different fibre volume fraction or constituent properties, requiring the usage of non-physical fitting parameter. In addition, extrapolating current knowledge to different loading/environmental conditions and to different manufacturing process will lead to a massive investment of resources.

On the other hand, it is well established that the mechanical properties of FRPs at the scale of mm are controlled by the constituent properties and spatial distribution of reinforcements. This circumstance leads naturally to a *bottom-up* strategy which is aimed to predict failure of composite materials and their structures directly from the constituents (fiber, matrix and their common interfaces), and moving up to the scale of a single ply, laminate, components and, finally, the whole structure (Figure 1.2). Although, micromechanical models have always been considered superior to purely phenomenological ones both in terms of accuracy and predictive capabilities, the actual development of robust micromechanical models has been hindered by two facts, namely the lack of accurate data about the mechanical properties of phases and interfaces at the nm-mm scale and the complexity of the three-dimensional (3D) microstructure, which precludes the use of simple analytic approximations in most cases. Nevertheless, the exponential growth of computational power in the last decades as well as the development of experimental techniques to characterize the mechanical behavior of materials at the nm-mm scale (mainly instrumented nanoindentation) have overcome these limitations.

Models based on computational micromechanics are very popular to understand the deformation and fracture mechanisms of engineering materials and to optimize the microstructure. In the case of UD FRPs composites, they have demonstrated

Figure 1.2: Bottom-up multiscale approach and virtual testing pyramid

their accuracy to predict the mechanical behavior as well as the fracture mechanisms under different loading conditions [González and LLorca \(2007a\)](#); [Totry et al. \(2009, 2010\)](#); [Vaughan and McCarthy \(2011b,a\)](#); [Canal et al. \(2012b,a\)](#), to determine the failure surface under multiaxial loading ([Totry et al. \(2008a,b,c\)](#); [Canal et al. \(2009\)](#); [Romanowicz \(2012\)](#); [Melro et al. \(2013b\)](#) or in the presence of porosity ([Ashouri Vajari et al. \(2014\)](#)), and as key elements within multiscale modeling strategies to carry out virtual testing of composite structures ([LLorca et al. \(2011, 2013\)](#)).

This new multiscale strategy takes a clear advantage from the composite structure, identifying three different entities (ply, laminate and component) whose mechanical behavior is characterized by three different length scales, namely fiber diameter, ply and laminate thickness, respectively. Fiber diameters are of the order

1.2. Multiscale modeling: towards virtual testing

of $5 - 20 \mu\text{m}$, while ply thicknesses are in the range $100 - 300 \mu\text{m}$ and standard laminates are several mm in thickness and above. The multiscale strategy is done by homogenizing the response of one entity at the corresponding length scale into a constitutive model that is passed to the next level.

Unidirectional plies are the building blocks of the multidirectional laminates used in the majority of structural applications. They can be modeled as transversely isotropic solids, whose elastic behavior is determined by five elastic constants: the elastic modulus in the fiber direction, E_1 , the elastic modulus perpendicular to the fibers, E_2 , the in-plane Poisson's ratio, ν_{12} , and the in-plane and out-of-plane shear moduli, G_{12} and G_{23} , respectively. They can be easily obtained from the elastic properties of the matrix and the fibers by means of simple micromechanical or analytic homogenization models (Böhm (2008)). Upon loading, unidirectional plies present a linear behavior until a critical stress is reached, leading to the onset of damage. Damage progresses rapidly afterwards and failure takes place at different manners depending on the loading condition.

Assuming that continuum mechanics describes the behavior even for the smaller scales (constituents), the finite element (FE) method can be used to homogenized the desired scale behavior prior to move to the upper one. In the first level, computational micromechanics is used to predict the ply properties from the thermo-mechanical properties of the constituents (fiber, matrix and their common interface), together with the volume fraction and spatial distribution of the fibers within an individual ply. Fiber properties (stiffness, strength, coefficients of thermal expansion) are usually provided by the material supplier. However, matrix and interface properties depend on the consolidation process (time, pressure and temperature, effect of fiber dispersion) and have a non-linear behavior that has to be characterized *in situ*.

Computational micromechanics is able to capture the onset and propagation of damage and to provide the magnitudes of strength and toughness associated to the different failure modes presented at the ply level. Once the homogenized ply properties are obtained, the information is passed to the laminate level and used into mesomechanical models, together with the stacking sequence and the inter-ply behavior, to determine the homogenized response of the laminate. Finally, the laminate information is used within the framework of computational mechanics to obtain the response until fracture of structural components, achieving the final aim of this multiscale approach, the *virtual testing* of composites (Figure 1.2).

1.3 Environmental effects on the mechanical response of FRPs

Among the different environmental agents affecting structures made of FRPs, those who can modify the temperature and the relative humidity of the composite require special attention. Temperature of the composite can be changed by any of the three heat transfer mechanisms, namely, conduction, convection and radiation. A composite structure in contact with hot parts (engine, exhaust pipes, etc) will act as heat sink and hence increase its temperature. However, since most polymers will melt at low temperature, this situation should be avoided (normally adding some isolating material). Therefore, convection and radiation stands as the main heat transfer mechanisms affecting the composite. Regarding convection, common examples are found in pipes, where the composite material is in permanent contact with a fluid that is at a certain temperature. Something similar happens to the fan cowls that are affected by the exhaust gases propelled by a jet engine nozzle. Regarding radiation, it can be found in large structures such as aircraft wings and fuselage directly heated by the Sun beams or small cavities affected by radiation coming from incandescent parts. Regarding the relative humidity, since water is everywhere, composite will inevitably absorb it. Of course, this situation is specially important in pipes and reservoirs (swimming-pools). Nevertheless, large aircraft structures such as wings can be affected as well by condensation of water by exceeding the saturation vapor pressure during flight (in service conditions).

Increasing the temperature of the FRP will lead to a strength and stiffness reduction mainly due to the presence of the polymer matrix. However, the fiber/matrix interface will be damaged too because of the stress concentration originated due to the mismatch of thermomechanical properties between the ceramic fibers and the polymer matrix.

Regarding the increase of the relative humidity, the water uptake will affect the composite putting down the mechanical properties of its constituents (mainly the polymer matrix and the fiber/matrix interface). Moisture will attack chemically (hydrolysis) the polymer molecules and the fiber/matrix interfacial bonds. In addition, water uptake is concentrated on the polymer, which will swell while fibers remain unaltered, provoking stress concentration at the fiber/matrix interfaces and leading to a significant decrease of the composite performance. Finally, not only the mechanical behavior but also the chemical structure of the polymer will be affected

1.4. Damage mechanisms and failure criteria in UD FRPs

by the presence of water, leading to changes on its internal structure and affecting its chemical properties (i.e changing the glass transition temperature, T_g). Water uptake is a complex process in fibrous composites, since its diffusivity depends on the constituents, their relative volume fractions, arrangement and morphology.

Although, the composite is affected by temperature and humidity separately, their effect is amplified when they act together. Larger temperatures induce larger water diffusion accelerating the degradation process. In addition, higher temperature during hygrothermal ageing not only increases the moisture uptake rate but it may also modify the local stress threshold required for delamination nucleation [Ray \(2006\)](#).

In view of this, a deeper knowledge about the HOT/WET behavior of the material and its influence in the structural design is mandatory. In aeronautics, the standard procedure to tackle the problem is through the so called HOT/WET knock down factors. To this end, the aircraft structure is assumed to be fully saturated (according to the level of relative humidity present in the ambient where the material is supposed to work, usually 85%) when the plane is under service. In addition, the structure is assumed to be operating at elevated temperature, which represents the worst scenario for the composite component. Under this conditions, the mechanical response of the composite is tested, subjecting coupons, previously aged according to the desired HOT/WET conditions, to different loading states. This process has to be repeated for sample belonging to the different levels of [Figure 1.2](#), increasing the time and costs as we climb the pyramid.

Understanding the combined effect of temperature and water absorption in FRPs from the constituent level provides an efficient methodology in order to estimate the performance of a UD FRP ply following a proper multiscale strategy, based on computational micromechanics. However, to this end a complete description of the constituent behavior under particular environments is required.

1.4 Damage mechanisms and failure criteria in UD FRPs

Despite all existing information and current knowledge about UD FRPs, the accurate prediction of the failure stress of composite materials and structures has been an elusive task due to the complexity of the failure mechanisms involved. Due to the anisotropy of the plies, six different physical uniaxial failure mechanisms can be identified in UD FRPs plies depending on the loading direction ([Figure 1.3](#)). The

tensile strength of the ply loaded parallel to the fibers, X_T , is controlled by the brittle fracture of the fibers, whereas the compressive strength along the same orientation, X_C , is dictated by the development of fibre kinking (Budiansky and Fleck (1995)). If the ply is subjected to stresses perpendicular to the fibers, the tensile strength Y_T is governed interface failure (Canal et al. (2012b,a)), and the compressive strength perpendicular to the fibers, Y_C , is dominated by the formation of a shear band of localized plastic deformation in the matrix across the ply (González and LLorca (2007a)). Finally, the shear strengths parallel and perpendicular to the fibers, S_L and S_T , are controlled by the matrix and interface shear strength.

However, these failure mechanisms are linked to each other, i.e., the compressive strength parallel to the fibers is severely reduced in the presence of shear stresses that facilitate the formation of kink bands (Vogler and Kyriakides (2001)). Thus, the maximum load-bearing capability of the ply under any stress state is dictated by a failure locus in the stress space. This failure locus (or failure criterion) is made up of the intersection of the various smooth surfaces, which correspond to the different physical failure modes depicted in Figure 1.3. In order to reduce the manufacturing time and the costs of new components, there is a need to establish the level of confidence in the methods for failure prediction of FRPs. To this end, Hinton et al. (2004b) carried out the worldwide failure exercise (WWFE), assessing nineteen different theoretical approaches for predicting the deformation and failure response of FRPs. The experimental results for the strength of the FRPs subjected

Figure 1.3: Schematic of the different failure micromechanisms in unidirectional plies as a function of the loading conditions

1.4. Damage mechanisms and failure criteria in UD FRPs

to complex stress states showed significant differences with the predictions of many theories, even when analyzing simple laminates.

The first failure criterion including damage propagation in UD laminates was proposed by Hashin in 1980 [Hashin \(1980\)](#), who considered two different failure mechanisms, one for the fiber and one for the matrix, assuming a quadratic interaction between normal and shear traction on the failure plane. Hashin pointed out that a failure criterion cannot be smooth but piecewise smooth, consisting of smooth branches each of which models a distinct failure mode. In addition, he suggested that identifying the failure mode (matrix tension, matrix compression, fiber tension or fiber compression) was crucial to capture not only the damage initiation but also its propagation. Unfortunately, Hashin's criterion was not able to determine the orientation of the failure plane for matrix and fiber compression failure modes observed in the experiments [González and LLorca \(2007a\)](#); [Kyriakides et al. \(1995\)](#). Moreover, the criterion did not capture the increase in shear strength due to compressive stress experimentally observed for matrix failure modes.

Puck and Schürman ([Puck and Schürmann, 1998](#)) proposed a modified version of Hashin's failure criterion that was able to capture the evolution of the angle of the fracture plane under matrix failure modes. Puck's stated that under matrix failure modes, fracture is caused by the combination of normal and shear stresses (σ_N , τ_T and τ_L) acting on a plane parallel to the fibers that he called the action plane (Figure 1.4).

The criterion distinguishes between three different failure modes (A, B and C)

Figure 1.4: Schematic of stresses acting on a parallel-to-fibre fracture plane inclined by the angle θ_{fp} against the thickness direction $\vec{e}_3$ (Puck's action plane).

Figure 1.5: Schematic of material failure envelope predicted by Puck under matrix failure modes (combination of transverse and in-plane stresses $\sigma_2 - \tau_{21}$).

for matrix failure modes. For matrix tension, the criterion predicts a failure plane parallel to the fibers and perpendicular to the loading direction f , whereas, under matrix compression the fracture plane was inclined at an angle of θ_{fp} ranging between 0° and 54° depending on the in-plane shear (Figure 1.5). Actually, under transverse compression, the set of Puck failure criteria for plane stress cases can be described by a single criterion:

$$\left(\frac{\tau_T}{R_{\perp\perp} - p_{\perp\perp}^c \sigma_N} \right)^2 + \left(\frac{\tau_L}{R_{\perp\parallel} - p_{\perp\parallel}^c \sigma_N} \right)^2 = 1 \quad (1.1)$$

, where $R_{\perp\parallel}$ and $R_{\perp\perp}$ are, respectively, the material shear strengths in fiber and transverse-to-fiber directions whereas $p_{\perp\parallel}^c$ and $p_{\perp\perp}^c$ are non-physical and experimentally-determined coefficients acting on the fracture plane. Therefore, Puck's criterion accounts for the influence of transverse compression on matrix shear strength by increasing the shear strength by terms that are proportional to the normal stress σ_N acting at the fracture plane.

Although Puck's criterion achieved a great success in the WWFE, it still presents some limitations, specially when dealing with longitudinal compression loading (fiber compression failure). In addition, Puck's failure criterion uses non-physical material parameters (such as $p_{\perp\parallel}^c$, $p_{\perp\perp}^c$ and $p_{\perp\parallel}^t$ shown in Figure 1.5) that may be difficult to determine for non-conventional material systems.

In order to overcome this limitations, a new set of six different phenomenological

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Figure 1.6: Schematic of LaRC principal failure modes.

failure criteria named as LaRC02/03 (Langley Research Center) was proposed by Dávila et al. and completed by Camanho et al. from 2002 to 2005 (Dávila et al. (2003); Camanho et al. (2003); Dávila et al. (2005)). The LaRC distinguishes four types of failure

In total, the LaRC distinguishes four different failure modes (Figure 1.6). The longitudinal tensile and compressive failure, are assumed to be fiber dependent modes, whereas the transverse failure modes are controlled by the matrix. These criteria are able to predict the angle of the fracture plane under matrix compression (transverse compression loading) as well as Puck's criteria do. Nevertheless, the criteria are also able to deal effectively longitudinal compression loading including the fiber kinking failure mechanism, which is predicted by calculating the fiber misalignment under load and applying the matrix failure criterion in the coordinate frame of the misalignment. Originally formulated for plane stress states, the LaRC predicts effectively the transition between the different failure modes and hence, the failure envelope (Figure 1.7). In addition, instead of using non-physical material parameters, the LaRC failure criteria obtain the fracture angle using material properties. In particular, using the Mohr-Coulomb failure criterion to model the ply behavior under transverse loading and maximizing the effective shear stresses respect to the angle of the failure plane.

The criteria were extended to account for in-plane shear non-linear behavior by Pinho et al. (Pinho et al. (2005)) and denoted as LaRC04. Recently, the model was combined with a pressure-dependent three-dimensional constitutive law (Pinho et al., 2012) to predict triaxial failure envelopes for different laminated composites included in the second World-Wide Failure Exercise.

Although both Puck and LaRC failure criteria are able to handle the different

matrix and fiber failure modes, the failure locus are obtained by means of phenomenological or physically based parameters, whose accuracy has to be verified through costly and time-consuming experimental campaigns (Puck et al. (2002); Dávila et al. (2005); Hinton et al. (2004a)). In addition, results obtained for a unidirectional ply could not be extrapolated to materials with different fibre volume fraction or constituent properties. Finally, the failure of unidirectional plies also depends on the energy dissipated by each damage mode, which has to be measured by means of complex mechanical tests or estimated by comparison with experiments (Pinho et al. (2006a); Camanho et al. (2014); Catalanotti et al. (2014)).

Figure 1.7: Schematic of material failure envelope predicted by LaRC under plane stress states: $\sigma_1 - \tau_{21}$ and $\sigma_1 - \sigma_2$.

1.5 Motivation and aims of the work

Fiber Reinforced Polymers (FRPs) are nowadays extensively used in applications where good mechanical properties are required in combination with weight savings. However, despite all existing information and current knowledge about these materials, the accurate prediction of the failure stress of composite materials and structures has been an elusive task due to the complexity of the failure mechanisms involved.

Various phenomenological and physically-based models have been proposed, whose input parameters have to be obtained through costly and time-consuming experimental campaigns for each material system. [Hinton et al. \(2004a\)](#); [Daniel et al. \(2009\)](#). Results obtained for a given unidirectional FRP system can not be directly extrapolated directly to other configurations with different fibre volume fraction or constituent properties, leading to a massive investment for their physical characterization. This is the case of material qualification for the aeronautical industry. As it was exposed at the beginning of this chapter, the tardy certification process of new materials makes the aeronautical industry extremely conservative limiting the usage of innovative materials that can improve the performance of the aircraft. The whole process can last well over two years due to the required tests under different agreeing and environmental conditions. Therefore, the motivation of this work is contributing to settle the basis of new reliable and faster certification processes and to reduce drastically time and cost associated to the current material choice programs.

Computational micromechanics (based on Finite Elements Analysis) offers a novel approach to understand the deformation and fracture mechanisms in materials engineering. In the case of unidirectional fibre-reinforced composites, it has demonstrated high accuracy in the prediction of the mechanical behavior, including fracture mechanisms under complex multiaxial loading cases [Totry et al. \(2008b\)](#); [Romanowicz \(2012\)](#); [Melro et al. \(2013b\)](#). Numerical simulations of Representative Volume Elements (RVE's) of the composite microstructure are useful to predict homogenized lamina properties, in close agreement with experimental data [Canal et al. \(2012a\)](#), and to provide the necessary input data for mesomechanical analysis at the laminate level. In addition, computational simulation of micromechanical RVE's can be used to reproduce experimental stress conditions rather difficult to impose experimentally in laboratory, such as biaxial or triaxial stress states. Moreover, the influence of the microstructure and the constituents properties in the failure mecha-

nisms can be addressed by means of parametric studies. All these efforts can lead in the future to the development micromechanical-based failure criteria with physical soundness, a clear advance in the state-of-the-art, e.g. Puck [Puck and Schürmann \(2002\)](#), LaRC [Dávila et al. \(2005\)](#) and Catalanotti [Catalanotti et al. \(2013\)](#) models.

In view of this and following previous research works [Segurado and LLorca \(2002\)](#); [Vaughan and McCarthy \(2010\)](#), herein detailed information of the microstructure (fiber diameter distribution, volume fraction, fiber clusters and resin pockets) is captured and included in a computational model of a unidirectional lamina (UD). Several strategies to determine micromechanical parameters by fitting against experimental results at the ply level, rather than measuring them with independent tests at the micro level, were developed in the past [Pinho et al. \(2006a\)](#); [Melro et al. \(2013a\)](#). However, the work developed in this thesis is aimed to predict the five ply strengths of a UD FRP, namely longitudinal tension strength (X_T), longitudinal compression strength (Y_C), transverse tension strength (Y_T), transverse compression strength (Y_C), and longitudinal shear strength (S_L) using a multiscale bottom-up strategy based on computational micromechanics and supported by micromechanical tests performed at the constituent level (fiber, matrix and fiber/matrix interface) under different environmental conditions (RT/DRY and HOT/WET). This bottom-up multi-scale simulation approach might lead in the future to a drastic reduction of the current costs associated with properties screening and material characterization programs [LLorca et al. \(2011\)](#).

In order to asses the validity of the computational models developed in this work, the ply strength predictions are compared to the values available in the literature. In addition, the RVE is submitted to multiaxial homogeneous stress states to determine the material failure envelope in the $\sigma_{22} - \tau_{12}$ plane under different environmental conditions. The model shows the importance of capturing adequately the competition between the different failure mechanisms, fiber/matrix debonding and matrix failure, operating at the same time when the material is subjected to mechanical and environmental loads.

1.6 Structure of this dissertation

Regarding the structure of this document, the introductory sections, already mentioned in this chapter, are followed by a description of the computational micromechanics framework, i.e. of the constitutive equations used to simulate matrix, fiber and interfaces, as well of the RVE generation procedure and the subsequent construction of the FE models with the specific loading conditions (chapter 2).

The remaining part of the work can be divided in two main parts, namely, experimental micromechanics and computational micromechanics. The former is gathered in chapter 3, which is devoted to describe the material system selected in this work as well as the different micromechanical techniques used to characterize the main properties of ply constituents.

In chapter 4, the computational micromechanical model used to predict the transversal behavior of unidirectional fiber reinforced polymers is explained. Whereas the chapter 5 is focused on the longitudinal behavior of the composite. In both chapters, the numerical predictions are compared against literature results.

Finally, chapter 6 is dedicated to multiaxial stress states and how to link the outputs from the micromechanical models to the mesomechanical scale. Last but not least, chapter 7 contains the main conclusions and some new developments proposed as future work.

Micromechanics for FRP composite materials

2

Continuum mechanics provides a suitable theoretical framework for continuous and homogeneous materials, fields vary smoothly over the neighborhood of a given material point. Moving downwards in the hierarchical structure of FRPs (see Figure 1.2), one can realize how considering the fiber and polymer matrix as the building blocks of the composite seems to be the first step along the multiscale bottom-up approach. These microconstituents, namely the fiber, the polymer matrix and its common interfaces represent the smallest FRPs entities described by continuum mechanics.

In order to study the micromechanical response of a composite, first, a Representative Volume Element (RVE) must be defined. The RVE represents the minimum material volume over in which a given property yields a value that is statistically representative of the overall one. Once this RVE size has been defined, the effective or macroscopical response of the composite can be obtained from the mechanical properties of the individual microconstituents. This computational homogenization process of the thermo-mechanical properties of the composite together with the development of new constitutive equations to study the damage onset and propagation are the main topics covered by micromechanics of FRPs.

In order to accomplish such a complex task, several micromechanical methods have been developed during the last years. [Böhm \(2008\)](#), classified them in two large branches, analytical and computational homogenization. A brief discussion of both micromechanical methods is presented in the next two sections.

2.1 Analytical micromechanics

Among the analytical methods, two different but closely related topics should be highlighted, the *mean field approaches* and the *variational bounds*. A brief description of both is outlined along the next paragraphs.

2.1.1 Mean-field approaches

These family of methods consider that the stress and strain fields for each constituent of the composite material can be represented by means of their corresponding volumetric averages. In the case of two-phased materials, such as FRPs, these fields can be named as $\bar{\sigma}_m$ and $\bar{\epsilon}_m$ for the matrix and $\bar{\sigma}_f$ and $\bar{\epsilon}_f$ for the fibers. Equation 2.1, shows the expression of the stress and strain fields averages over the volume of each individual phase, where $i=m,f$, $V = V_m + V_f$ is the total volume and $\mathbf{x}$ is the position vector.

$$\bar{\sigma}_i = \frac{1}{V_i} \int_{V_i} \sigma_i(\mathbf{x}) dV \quad (2.1a)$$

$$\bar{\epsilon}_i = \frac{1}{V_i} \int_{V_i} \epsilon_i(\mathbf{x}) dV \quad (2.1b)$$

Assuming elastic behavior for both phases, stresses and strains can be related using the fourth-order stiffness tensor $\mathbf{L}$. The average stress field $\bar{\sigma}$ over the volume of the RVE is shown in Equation 2.2, where ξ corresponds to the fiber volume fraction

$$\bar{\sigma} = (1 - \xi)\bar{\sigma}_m + \xi\bar{\sigma}_f = (1 - \xi)\mathbf{L}_m\bar{\epsilon}_m + \xi\mathbf{L}_f\bar{\epsilon}_f \quad (2.2)$$

On the other hand, mean fields of stress and strain on each phase ($i=m,f$) can be determined with the overall ones $\bar{\sigma}$ and $\bar{\epsilon}$ through the fourth order concentration tensors $\mathbf{A}_i$ and $\mathbf{B}_i$, as

$$\bar{\epsilon}_i = \mathbf{A}_i\bar{\epsilon} \quad (2.3a)$$

$$\bar{\sigma}_i = \mathbf{B}_i\bar{\sigma} \quad (2.3b)$$

2. Micromechanics for FRP composite materials

These set of stress and strain fourth order concentration tensors should fulfill the volume average over the RVE as

$$(1 - \xi)\mathbf{A}_m + \xi\mathbf{A}_f = \mathbf{I} \quad (2.4a)$$

$$(1 - \xi)\mathbf{B}_m + \xi\mathbf{B}_f = \mathbf{I} \quad (2.4b)$$

where $\mathbf{I}$ is the fourth-order identity tensor. Finally, if the matrix stress and strain fields are replaced in Equation 2.2 then the overall stiffness and compliance of the homogenized material can be determined as

$$\mathbf{L} = \mathbf{L}_m + \xi(\mathbf{L}_f - \mathbf{L}_m)\mathbf{A}_f \quad (2.5a)$$

$$\mathbf{M} = \mathbf{M}_m + \xi(\mathbf{M}_f - \mathbf{M}_m)\mathbf{B}_f \quad (2.5b)$$

Therefore, the elastic problem in mean fields consists on determining one of these four concentration tensors, $\mathbf{A}_m$, $\mathbf{A}_f$, $\mathbf{B}_m$ or $\mathbf{B}_f$, for a particular RVE. $\mathbf{A}$ and $\mathbf{B}$ depend on the volume fraction of the constituents and on the shape and spatial distribution of the reinforcement phase. These methods are also known as estimation methods.

Eshelby (1957), determined average strain produced in an ellipsoidal inclusion embedded in an infinite media, determining the exact solution for the strain concentration tensor. As that depends only on the Poisson ratio of the matrix and on the ellipsoidal shape. The tensor $\mathbf{S}_f$, named as the Eshelby's tensor, includes the effect of the inclusion used (ellipsoids, discs, long fibers, etc.). If the strain concentration tensor $\mathbf{A}_f^{\text{dil}}$ is determined, then the elastic constants of the heterogeneous material can be obtained as

$$\mathbf{L} = \mathbf{L}_m + \xi(\mathbf{L}_f - \mathbf{L}_m)[\mathbf{I} + \mathbf{S}_f\mathbf{L}_m^{-1}(\mathbf{L}_f - \mathbf{L}_m)]^{-1} \quad (2.6)$$

This expression gives good predictions under dilute conditions, where the inclusion volume fraction is below 10%.

In order to overcome this limitation, new micromechanical models were developed varying the procedure to estimate the stress and strain concentration tensors.

2.1. Analytical micromechanics

Among all the models, two of them have been very successful and should be highlighted in this work. In chronological order, the classical self consistent (CSC) model (Kröner (1958)) and the Mori-Tanaka (MT) model Mori and Tanaka (1973); Benveniste (1987).

In the CSC model, the concentration tensor is calculated assuming an inclusion embedded in a composite material with unknown properties, that are indeed the solution of the problem. To do so, the Eshelby's concentration tensor, $\mathbf{A}_f^{\text{dil}}$, is modified substituting the matrix stiffness tensor, $\mathbf{L}_m$, by the unknown composite one $\mathbf{L}$ and obtaining a new concentration tensor $\mathbf{A}_f^{\text{sc}}$ (Equation 2.7). Therefore, substituting $\mathbf{A}_f^{\text{sc}}$ in the general expression 2.5 results in a non-linear equation (Equation 2.8) that can be solved by fixed point iterations. This model is especially suited for polycrystals, where there is no continuous matrix reinforced by particles or fibers.

$$\mathbf{A}_f^{\text{sc}} = [\mathbf{I} + \mathbf{S}_f \mathbf{L}^{-1} (\mathbf{L}_f - \mathbf{L}_m)]^{-1} \quad (2.7)$$

$$\mathbf{L} = \mathbf{L}_m + \xi (\mathbf{L}_f - \mathbf{L}_m) [\mathbf{I} + \mathbf{S}_f \mathbf{L}^{-1} (\mathbf{L}_f - \mathbf{L}_m)]^{-1} \quad (2.8)$$

On the other hand, the MT model estimates the concentration tensor $\mathbf{A}_f^{\text{mt}}$ by interpolating it between the two bounds. First, if $\xi \rightarrow 0$, the strain concentration tensor corresponds to the Eshelby's dilute concentration tensor $\mathbf{A}_f^{\text{dil}}$ while if $\xi \rightarrow 1$ the identity tensor $\mathbf{I}$ (Equation 2.9) should be recovered. Therefore the overall stiffness tensor of the homogenized material can be directly obtained through Equation 2.10.

$$\mathbf{A}_f^{\text{mt}} = \mathbf{A}_f^{\text{dil}} [(1 - \xi) \mathbf{I} + \xi \mathbf{A}_f^{\text{dil}}]^{-1} \quad (2.9)$$

$$\mathbf{L} = \mathbf{L}_m + \xi (\mathbf{L}_f - \mathbf{L}_m) [\mathbf{A}_f^{\text{dil}} [(1 - \xi) \mathbf{I} + \xi \mathbf{A}_f^{\text{dil}}]^{-1}] \quad (2.10)$$

An important modification of the SC method, named as the generalized self consistent (GSC) model was developed by Christensen and Lo (1979) in order to deal with elastic matrix reinforced with long fibers or spherical particles. The GSC provides better results than the MT method in the prediction of the elastic properties of spherical-reinforced composites. However, since the GSC is not a proper mean field method is not explained here. More details can be found in Christensen (1990).

2.1.2 Variational bounding methods

These methods use variational principles, based on the principle of a minimum potential energy, in order to bound the mechanical properties of heterogeneous materials. Perhaps the most well-known bounds are the Hill's limits (Equation 2.11), which comprise the isostrain Voigt lower bound and the isostress Reuss upper bound. This variational method only considers the volume fraction of the reinforcements as microstructural information. Neither their shape nor their distribution are taken into account, providing worthless predictive capabilities out of the isostrain or isostress loading conditions.

$$[\xi \mathbf{M}_f + (1 - \xi) \mathbf{M}_m]^{-1} \leq \mathbf{L} \leq \xi \mathbf{L}_f + (1 - \xi) \mathbf{L}_m \quad (2.11)$$

Hashin and Strikman [Hashin and Shtrikman \(1963\)](#) proposed a variational method for statistically isotropic microstructures. This variational method narrowed the Hill's limits providing tighter bounds due to the inclusion of additional information if the microstructure.

Finally, there is another family of variational bounding methods, known as the three-point limits, which use the reinforcement volume fraction, ξ , as well as statistical information about the distribution of the different phases in the composite material. This information is incorporated to the method by means of some micromechanical parameters admitting closed-form solutions for some particular cases, such as random dispersion of hard spherical particles with isotropic and homogeneous distributions. Among all the expressions available in the literature, it is worthy to mention the work developed by [Torquato \(1998\)](#), whose models for biphasic materials provide most probably the best analytical solution to calculate the elastic constants of composite materials reinforced with elastic inclusions.

2.2 Computational micromechanics

Extending the analytical models to the non-linear regime, including plasticity and damage, can be extremely inaccurate due to the impossibility to account for large strain gradients in mean field methods. In addition, including manufacturing conditions, such as thermal residual stresses and porosity, and dealing with anisotropic microstructures will make the problem almost unfeasible. Fortunately, computational homogenization by means of any numerical method (FE, DFT, etc), can account for

2.2. Computational micromechanics

complex non-linear behaviors, such as damage and plasticity as well as finite strains. In addition, the details of the stress and strain microfields are resolved, and thus it is possible to track the nucleation and growth of damage during the analysis, a key aspect to model the localization of damage and fracture. Finally, computational homogenization can deal with complex multiaxial stress states, difficult to achieve in laboratory conditions, to obtain the failure locus of the heterogeneous material. For these reasons and thanks to the rapid increase of computational resources, computational homogenization models are becoming more and more popular these days.

In order to simulate the behavior of a discrete microstructure, two different strategies can be adopted, the embedded cell method and the periodic microfield approach. The former is mainly used to model the evolution of the stress and strain field when damage is localized along a well defined region, whereas the latter is devoted to predict the effective response of the heterogeneous material up to the onset of damage propagation. The advantages and disadvantages as well as and the specific ranges of applicability of both methods are outlined along the next paragraphs.

2.2.1 Periodic RVE models

The first step in this homogenization strategy consists in the creation of a suitable statistically representative periodic RVE to approximate the mechanical response of an infinite composite lamina (Figure 2.1). In the case of a unidirectional fiber reinforced composite, the RVE is modeled as a random and homogeneous distribution of fibers periodically dispersed in the polymer matrix. Therefore, the challenge consists of generating these periodic fiber distributions. Sometimes, depending on the material, ideal microstructures where the fiber packing can be approximated using simplified fiber distributions (hexagonal, square, etc.), are enough. However, studying the damage mechanisms of FRPs requires, in most cases, complex microstructures whose generation relies on information extracted from real micrographs of the reinforcement distribution. Under these conditions, the size of the RVE and, hence, the number of fibers, should exceed a minimum value to contain all the necessary information about the statistical description of the microstructure. In addition, the RVE should be large enough so that the average properties of this volume element are independent of its size. Obviously, the critical size of the RVE depends on the phase and interface properties and spatial distribution. Usually, periodic distributions can be used to overcome layer effect problems that trigger damage at the

Figure 2.1: Schematic of periodic RVE (red) representing the inner region of a FRP unidirectional ply (blue)

boundaries of the RVE. In computational homogenization, increasing the size of the RVE will blow up the number of elements and degrees of freedom of the FE model, making the computation rather unfeasible. Fortunately, a few dozens of fibers are enough to capture the overall behavior of FRPs (Totry et al. (2010); González and LLorca (2007b)).

Virtual multi-fiber periodic microstructures generation

In order to generate real-like microstructures, fiber distributions can be directly determined from a micrograph of the composite cross-section, using image-based reconstruction techniques (Trias et al. (2006); Romanov et al. (2013)), or directly generated using mathematical algorithms (Rintoul and Torquato (1997); Segurado and LLorca (2002); Vaughan and McCarthy (2010); Melro et al. (2008)). Image-based reconstruction techniques offers the best RVE in terms of fidelity to the real microstructure, capturing the presence of defects, fiber clusters, etc. Although these methods are improving very fast, high quality composite cross-section micrographs are not always available. In addition, sample preparation, image processing and the export process to the FE code are tough tasks, requiring a large amount of time and computer resources compared to the solution provided by the algorithms. Finally,

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periodic microstructures are not easily found in real micrographs and enforcing such condition leads to an extra effort that will make the mathematical work unfeasible. Therefore generating a virtual microstructure statistically equivalent to the real one based on random distributions is still the faster methodology to tackle the problem.

Even in the case of random models, the generation of the periodic RVE is not straightforward, since increasing the number of fibers means increasing either the fiber volume fraction or the size of the RVE. Among the different alternatives, two algorithms stand out over the rest, the Random Sequential Adsorption (RSA) and the Nearest Neighbor Algorithm (NNA).

The RSA algorithm proved its robustness and offered a remarkable speed even for the generation of large 3D spherical particle distributions [Segurado and LLorca \(2002\)](#). The NNA considers additional statistics, such as the distribution of fibre diameters and fibre neighboring distances. As a consequence, the algorithm is able to reproduce distribution patterns, fiber clusters and matrix rich regions observed in the real micrographs. Since the NNA takes into account additional statistical information, the resulting RVEs are expected to provide better accuracy, in the FE simulations, compared to the ones generated by means of the RSA. Nevertheless, it was observed that the results obtained using both algorithms can be very similar in equivalent conditions [Naya et al. \(2014\)](#).

According to the original RSA algorithm ([Rintoul and Torquato \(1997\)](#)), fibers centers are generated randomly and sequentially, and each new fibre is accepted if the distance between neighboring fibre surfaces is higher than a minimum distance (of the order of $0.1R$, where R stands for the fibre radius) to ensure an adequate finite element discretization of this region. In addition, the distance between the fibre surface and the RVE edges should be higher than the same critical distance to avoid distorted finite elements during meshing. At this point [Segurado and LLorca \(2002\)](#) introduced a modification to increase the fiber volume fraction. Therefore, if the target fiber volume fraction is not achieved, the unit cell is compressed in several steps. Each fiber is adsorbed towards a certain point at every step, increasing the local volume fraction (Figure 2.2). If a fiber overlaps a neighbor, it returns to the previous position. The process finishes once the desired fibre volume fraction within the RVE is reached or if any fiber can no longer be moved.

Regarding the NNA, the generation process is based on the classification method proposed by Fix and Hodges ([Silverman and Jones \(1989\)](#)). As it was implemented by [Vaughan and McCarthy \(2010\)](#), the NNA is similar to the original RSA. However,

new fibers are added according to statistical measurements instead of randomly. The first fiber is placed in a random position inside the limits of the RVE, then a second fiber (larger or smaller) is added at a certain distance, which is not arbitrary. Both the diameter and the first neighbor distance follow an statistical distribution obtained from the analysis of a real micrograph of the composite cross-section. The process is repeated for new fibers(the third, the fourth and so on), which are placed according to their relative neighbor distance to the first fiber. Normally, the process is extended up to two consecutive fibers(the second and the third), hence, considering only the first and the second neighbor distances. Then, a new fiber(the third) is added in another random position and the process starts again (Figure 2.3). Of course, overlaps need to be checked for every new fiber.

Figure 2.2: Schematic of the periodic fiber distribution generation using the Random Sequential Adsorption (RSA) algorithm

Figure 2.3: Schematic of the periodic fiber distribution generation using Nearest Neighbor Algorithm (NNA)

Periodic boundary conditions

According to Hazanov and Huet (1994), periodic boundary conditions (PBC) offer a balance between the effective response obtained imposing displacements (upper limit) and tensions (lower limit). Although, the three types of boundary conditions will converge to the same result if the RVE is large enough, among them the PBC offer the best results for a given RVE size. Regarding 3D RVEs, the PBC are normally expressed in terms of three displacement vectors $\mathbf{u}_1$, $\mathbf{u}_2$ and $\mathbf{u}_3$ relating the relative displacement of a set of master nodes (typically M0, MX, MY and MZ) laying in opposite faces. Therefore $\mathbf{u}_1 = \mathbf{u}^{\text{MX}} - \mathbf{u}^{\text{M0}}$, $\mathbf{u}_2 = \mathbf{u}^{\text{MY}} - \mathbf{u}^{\text{M0}}$ and $\mathbf{u}_3 = \mathbf{u}^{\text{MZ}} - \mathbf{u}^{\text{M0}}$. The PBC ensure the continuity between neighboring RVE, which will deform like a jigsaw puzzle (Figure 2.4).

According to Figure 2.5, the PBC can be mathematically expressed as Equation 2.12.

$$\mathbf{u}(L, y, z) - \mathbf{u}(0, y, z) = \mathbf{u}^{\text{MX}} - \mathbf{u}^{\text{M0}} = \mathbf{u}_1 \quad (2.12a)$$

$$\mathbf{u}(x, H, z) - \mathbf{u}(x, 0, z) = \mathbf{u}^{\text{MY}} - \mathbf{u}^{\text{M0}} = \mathbf{u}_2 \quad (2.12b)$$

$$\mathbf{u}(x, y, W) - \mathbf{u}(x, y, 0) = \mathbf{u}^{\text{MZ}} - \mathbf{u}^{\text{M0}} = \mathbf{u}_3 \quad (2.12c)$$

The relative displacement of a couple of nodes laying on opposite faces is equal to the displacement between the corresponding pair of master nodes (Figure 2.5). The different loading states, comprising any physical homogeneous deformation state within the unit cell, are introduced imposing displacements to these master nodes. Each master node will contain the reaction force generated by all the nodes laying on its corresponding face. For example, a uniaxial tensile strain state along the x axis is introduced by setting $\mathbf{u}^{\text{MX}} = (\delta, 0, 0)^T$, $\mathbf{u}^{\text{MY}} = (0, u_y, 0)^T$ and $\mathbf{u}^{\text{MZ}} = (0, 0, u_z)^T$, where u_y and u_z are obtained from the homogenized traction acting on their corresponding faces $x = L$ and $y = H$ (Equation 2.13).

$$\int_{\Omega_y} t_y d\Omega = 0 \quad \Omega_y : y = H \quad (2.13a)$$

$$\int_{\Omega_z} t_z d\Omega = 0 \quad \Omega_z : z = W \quad (2.13b)$$

Figure 2.4: Schematic of the different failure micromechanisms in unidirectional plies as a function of the loading conditions

Figure 2.5: Schematic of the different failure micromechanisms in unidirectional plies as a function of the loading conditions

2.2.2 Embedded cell models

In these models, fracture process zone is approximated by means of a detailed microstructure of a real composite that is embedded in the effective material whose properties are those of the homogenized composite (Figure 2.6). This outer region is used to transfer the far-field loads, applied on the boundaries, to the microstructural core. Therefore, these models are not devoted to predict the effective response of the material but the evolution of the stress-strain fields and damage mechanisms within the fracture process zone. The main advantage of the embedded cells is that the periodicity of the microstructure is no longer required, simplifying the pre-processing tasks considerably.

Very often, three different strategies can be found in the literature depending on how the outer region is modeled Böhm (2008). The first option, both the core and the outer region are represented as an heterogeneous material providing a highly detailed description of the microstructure. However, even if a very coarse mesh is used in the outer region, this approximation becomes extremely expensive from a computational point of view. In order to avoid such a problem, especially in medium-large models, there is a second alternative where the outer region is modeled as an homogeneous material whose properties can be approximated using one of the mean-field methods described in the previous section. This new possibility is still expensive but less than the first one and is especially suited to study the localization and growth of cracks Canal et al. (2012b); González and LLorca (2007a). However, the transition between the core and the outer region is not always smooth. In FRPs, this strategy is normally limited to 2D intraply failure studies, unless very large FE models can be used. Finally, there is a third alternative consisting on using an homogeneous outer region with unknown properties and solving the problem in an iterative way, in a similar manner as for the self consistent method. However, extending this approach to the non-linear regime is still problematic. Therefore, the second option is considered as the better balance between predictive capabilities and computational requirements.

Figure 2.6: Schematic of an embedded cell simulation of fracture. The composite microstructure is considered modeling the constituents (matrix, reinforcements and interfaces) embedded in the homogeneous material (gray) whose constitutive equation is provided by a suitable homogenization model. The crack is highlighted in red.

2.3 Constitutive behavior of FRPs constituents

Although the micromechanical models based on FE represent explicitly the different constituents of the FRP, capturing the deformation and damage mechanisms is still a tough task. To this end, specific constitutive equations are employed to model each constituent behavior, namely fibers, matrix and fiber/matrix interface.

2.3.1 Matrix plasticity

Polymers are strain rate sensitivity solids. Although, as a first approximation, strain rate effects are usually neglected when dealing with quasistatic applications, modeling the behavior of polymers in the quasi-static regime is still a complex task.

Assuming that, for low strain rate applications, polymers are modeled as simple elasto-plastic solids, there are still major differences when polymers are compared to standard elasto-plastic solids, such as metals. Under tensile loading, polymers usually fail in a brittle manner at very low strain, whereas, under compression, they exhibit large plastic deformation (Fiedler et al., 2001). In addition, the yield behavior of polymers is pressure-sensitive, which means that the yield stress decreases with hydrostatic tension and increases with hydrostatic compression. In view of this, capturing the evolution of the yield stress requires a proper yield criterion such as the Mohr-Coulomb (Labuz and Zang (2012)) or the Drucker-Prager (Drucker and Prager (1952)). Originally developed to establish the yield behavior of soils and rocks, these pressure-dependent yield criteria have been adopted in the literature González and LLorca (2007b); Melro et al. (2013a); Yang et al. (2012); Ashouri Vajari et al. (2014); Canal et al. (2009); Fiedler et al. (2001). Both plasticity criteria are detailed along the next paragraphs.

Plasticity and tensor invariants

Although the yield criteria can be formulated in terms of Cauchy stress tensor components, σ_{ij} , it is very convenient to do it in terms of the principal stresses, σ_1, σ_2 and σ_3 . In fact, since the three principal stresses require an orientation with respect to the material directions, the yield criteria should be formulated in terms of tensor invariants. Very often, yield criteria are expressed as a function of I_1 , J_2 and J_3 , where I_1 is first Cauchy's stress tensor invariant and J_2 and J_3 are the non-zero principal invariants of the deviatoric stress tensor $s_{ij} = \sigma_{ij} - \frac{1}{3}\sigma_{kk}\delta_{ij}$. The expression of I_1 , J_2 and J_3 in terms of σ_{ij} and the principal stresses are gathered in Equation 2.14 and Equation 2.15, respectively, where s_1 , s_2 and s_3 stand for the principal deviatoric stresses.

$$I_1 = \sigma_{ii} \tag{2.14a}$$

$$J_2 = \frac{1}{2}s_{ij}s_{ij} \tag{2.14b}$$

$$J_3 = \frac{1}{3}s_{ij}s_{jk}s_{ki} \tag{2.14c}$$

2.3. Constitutive behavior of FRPs constituents

$$I_1 = \sigma_1 + \sigma_2 + \sigma_3 \quad (2.15a)$$

$$J_2 = \frac{1}{6} [(\sigma_1 - \sigma_2)^2 + (\sigma_1 - \sigma_3)^2 + (\sigma_2 - \sigma_3)^2] \quad (2.15b)$$

$$J_3 = s_1 s_2 s_3 \quad (2.15c)$$

Looking at the expression of J_2 , the von Mises equivalent stress σ_y^{VM} is easily recovered, providing a compact expression for the von Mises yield criteria (Equation 2.16) and relating J_2 to the magnitude of the yield limit.

$$f(J_2) = \sqrt{J_2} - \tau_y^{\text{VM}} = \sqrt{J_2} - \frac{\sigma_y^{\text{VM}}}{\sqrt{3}} = 0 \quad (2.16)$$

In order to provide a geometrical interpretation of the yield surface, the three principal stresses can be used as the base of a three dimensional space. Therefore, the line placed at equal angles respect to all three principal axes defines a space diagonal, where $\sigma_1 = \sigma_2 = \sigma_3$ and hence, points on it are in a state of hydrostatic stress, which means that $\sigma_{ij} = \sigma_m \delta_{ij}$, where $\sigma_m = \sigma_{ii}/3 = I_1/3$ stands for the hydrostatic stress.

The geometrical interpretation of I_1 in the formulation of the yield surface is clear thanks to the definition of the hydrostatic axis. However, the visualization of J_2 and J_3 requires the definition of another geometrical entity, the π -plane. Among the infinity number of planes perpendicular to the hydrostatic axis, there is one of them, the π -plane, where $\sigma_1 + \sigma_2 + \sigma_3 = 0$. Therefore, if the yield criteria are independent of I_1 invariant (i.e von Mises and Tresca criteria), the stress state at any point, represented by the vector $\boldsymbol{\sigma} = (\sigma_1, \sigma_2, \sigma_3)$, is the sum of the stress state at the corresponding point of the π -plane, represented by the deviatoric stress vector $\boldsymbol{\rho} = (s_1, s_2, s_3)$, and the hydrostatic stress vector $\boldsymbol{\xi} = (\sigma_m, \sigma_m, \sigma_m)$ (Figure 2.7). Moreover, the yield locus at this plane can be defined as a function of the module and the direction of the vector $\boldsymbol{\rho} = (s_1, s_2, s_3)$, which are related to the J_2 and the J_3 invariants respectively.

However, to visualize the pressure-dependent yield criteria commonly used to represent the plastic behavior of polymers, it is interesting define a new system of cylindrical coordinates (ξ, ρ, Θ) called the Haigh-Westergard space. The first two coordinates of this new system have been already defined thanks to the hydrostatic and deviatoric stress vectors, $\boldsymbol{\xi}$ and $\boldsymbol{\rho}$. The third component of the Haigh-Westergard

space is the Lode angle Θ , which accounts for the variation of ρ on the π -plane and contains the information provided by the J_2 and the J_3 invariants. The Lode angle accounts for the influence of the middle principal stress and ranges between $-\pi/6$ (compression meridian) and $\pi/6$ (tension meridian). It vanishes at pure shear (Figure 2.7). Mathematically, the three coordinates ξ , ρ and Θ can be expressed as a function of the invariants I_1 , J_2 and J_3 as

$$\xi = |\boldsymbol{\xi}| = \sqrt{3}\sigma_m = I_1/\sqrt{3} \quad (2.17a)$$

$$\rho = |\boldsymbol{\rho}| = \sqrt{2J_2} \quad (2.17b)$$

$$\cos 3\Theta = \frac{3\sqrt{3}}{2} \frac{J_3}{J_2^{3/2}} \quad (2.17c)$$

The relationship between the Haigh-Westergard coordinates and the principal stresses σ_1, σ_2 and σ_3 is gathered in Equation 2.18.

$$\begin{bmatrix} \sigma_1 \\ \sigma_2 \\ \sigma_3 \end{bmatrix} = \frac{1}{\sqrt{3}} \begin{bmatrix} \xi \\ \xi \\ \xi \end{bmatrix} + \sqrt{\frac{2}{3}} \rho \begin{bmatrix} \cos \Theta \\ \cos \Theta - \frac{2}{3}\pi \\ \cos \Theta + \frac{2}{3}\pi \end{bmatrix} \quad (2.18)$$

In view of this, the yield surface can be defined according to its intersection with two different planes: the previously mentioned π -plane and the so called meridian plane, which is defined by the $\boldsymbol{\xi}$ and $\boldsymbol{\rho}$ vectors (Figure 2.7). This two geometrical entities will provide a better understanding of the most common pressure-dependent yield functions used to model plasticity of polymers, the Mohr-Coulomb and the Drucker-Prager criteria. Both of them are explained along the next sections.

Mohr-Coulomb failure criterion

The Mohr-Coulomb failure criterion relates the shear stress required to produce material yielding (τ_y) to the applied normal stress by means of a linear relationship that can be mathematically expressed as Equation 2.19. The model is based on Coulomb's friction hypothesis and relies on Mohr's theory to determine the combination of shear and normal stresses producing the material failure. The angle ϕ represents the material internal friction and it accounts for the influence of the normal stress σ_n in the evolution of the yield surface. The parameter c is called the material cohesion and corresponds to the intersection of the failure envelope with

2.3. Constitutive behavior of FRPs constituents

Figure 2.7: Schematic of the transformation from the principal stress space to the Haigh-Westergard space

the τ axis. Therefore the failure envelope and hence the plastic behavior of the polymer is defined by the material cohesion c and the internal friction angle ϕ .

$$\tau_y = c - \sigma_n \tan \phi \quad (2.19)$$

In terms of principal stresses, σ_1 , σ_2 and σ_3 , the Mohr-Coulomb criterion can be expressed as Equation 2.20. In view of this expression, the material compression and tension yield stress σ_{yc} and σ_{yt} , respectively, can be expressed as Equation 2.21.

$$\sigma_1 - \sigma_3 = 2c \cos \phi - (\sigma_1 + \sigma_3) \sin \phi \quad (2.20)$$

$$\sigma_{yt} = 2c \frac{\cos \phi}{1 + \sin \phi} \quad (2.21a)$$

$$\sigma_{yc} = 2c \frac{\cos \phi}{1 - \sin \phi} \quad (2.21b)$$

Under uniaxial compression, the shear band inclination plane of a solid, which follows the Mohr-Coulomb criterion, forms an angle θ , respect to plane perpendicular

to the loading axis, that can be related to ϕ according to $\theta = \pi/4 + \phi/2$ (Figure 2.8).

Figure 2.8: Schematic of the Mohr-Coulomb failure criterion

In terms of the Haigh-Westergaard coordinates, the Mohr-Coulomb yield criterion can be expressed according to Equation 2.22.

$$f(\xi, \rho, \Theta) = \sqrt{2} \xi \sin \phi + \sqrt{3} \rho \sin(\Theta + \frac{\pi}{3}) + \rho \cos(\Theta + \frac{\pi}{3}) \sin \phi - \sqrt{6} c \cos \phi = 0 \quad (2.22)$$

In view of this, the failure locus on the π -plane and the meridian plane can be constructed, as it is displayed in Figure 2.9a. The failure locus on the π -plane has the shape of an irregular hexagon, whereas in the meridian plane it looks like an irregular cone that cuts the ξ axis at $\xi = \sqrt{3}c/\tan \phi$.

In view of this, the Mohr-Coulomb yield surface in the space defined by the principal stresses is an irregular hexagonal pyramid (Figure 2.10) that clearly shows the dependency of this yield criterion with respect to the hydrostatic pressure.

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Figure 2.9: Schematic of the Mohr-Coulomb failure criterion on the Haigh-Westergaard deviatoric and meridian planes. Compression and tension meridians are highlighted in red and blue respectively

It should be noticed that when $\phi = 0$ the Mohr-Coulomb model reduces to the pressure-independent Tresca model while $\phi = \pi/2$ leads to *tension cut-off* Rankine model (Rankine, 1856).

Figure 2.10: Schematic of the Mohr-Coulomb yield surface in the three dimensional principal stress space. Compression and tension meridians are highlighted in red and blue respectively

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Drucker-Prager failure criterion

The Mohr-Coulomb criterion has been widely used to model the behavior of soils, and specially non-cohesive materials ($c \approx 0$) such as gravels and sands. However, it has several drawbacks. First, the lack of intermediate principal stress dependency, which can underestimate the strength of the material in many cases. On the other hand, the vertex found on its yield surface leads to numerical issues related to the evolution of the plastic flow. A common way to avoid such inconvenient consists of smoothing the yield surface, changing the evolution of the plastic flow in both the meridian and the deviatoric planes. Among the different failure criteria devoted to this end, perhaps the Drucker-Prager stands out due to its relative simplicity and ease matching the Mohr-Coulomb behavior.

The Drucker-Prager yield criterion (Drucker and Prager (1952)) has been successfully applied to model several pressure-dependent materials such as soils, rocks, concrete and polymers. This failure criterion is very often formulated in terms of shear and normal pressure ($p = -\sigma_n$) as it was done for the Mohr-Coulomb. The expression is gathered in Equation 2.23, where d is the material cohesion and β is the material internal friction angle. In view of this, the similarities between d and β with the Mohr-Coulomb c and ϕ parameters are evident. In fact, the Drucker-Prager, originally developed for soils, is often considered as a smooth version of the Mohr-Coulomb criterion.

$$\tau_y = d + p \tan \beta \quad (2.23)$$

However, despite its close relation to the Mohr-Coulomb, the Drucker-Prager yield criterion can be seen as an evolution of the popular von Mises criterion, including the effect of the hydrostatic pressure in the plastic evolution. In fact, in terms of the tensor invariants, I_1 and J_2 , the D-P can be expressed according to Equation 2.24, where the similarities with the von Mises criterion (Equation 2.16) are evident.

$$f(I_1, J_2) = \alpha I_1 + \sqrt{J_2} - d = 0 \quad (2.24)$$

where α is the pressure sensitivity parameter. Since $I_1 = 3p$, little algebra provides the equivalence between α and β through $3\alpha = \tan \beta$. If the D-P is expressed in terms of the Haigh-Westergaard coordinates, Equation 2.25, it can be observed how in the π -plane ($\xi = 0$) the D-P yield locus is a circle of radius

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$\rho = \sqrt{2}d$, in a similar manner as it is for the von Mises criterion. However, in the meridian plane $(\xi - \rho)$, the D-P appears as cone with apex at $d/\sqrt{3}\alpha$ (Figure 2.11).

$$f(\xi, \rho) = \sqrt{6}\alpha\xi + \rho - \sqrt{2}d = 0 \quad (2.25)$$

Since the yield locus on the π -plane does not depend on the lode angle Θ , matching the D-P and the M-C criteria has different possibilities depending on the chosen value of ρ (Figure 2.12). If the match is at $\Theta = -\pi/6$ (compressive meridian), the D-P cone circumscribes the M-C hexagonal pyramid. On the other hand, matching both surfaces at $\Theta = 0$ (tensile meridian) the D-P surface inscribes the M-C (Figure 2.11). The D-P can match as well the M-C at the shear meridian ($\Theta = 0$), but this option is perhaps the rarest one, because of the difficulties associated to the shear test.

Figure 2.11: Schematic of the Ducker-Prager failure surface on the Haigh-Westergaard deviatoric and meridian planes fitting the Mohr-Coulomb failure surface at the compression and tension meridians (highlighted in red and blue respectively)

Figure 2.12: Schematic of the Drucker-Prager yield surfaces (red and blue cones) in the three dimensional principal stress space fitting the Mohr-Coulomb pyramid at two different meridians (compression and tension respectively). The red cone circumscribes the pyramid where as the blue one is inscribed on it

2.3.2 Modeling damage at the microscale

Modeling damage inside a composite UD ply requires to understand the nature and origin of damage at the different constituents. In particular, the polymer matrix and the fiber/matrix interface are the weakest parts of the microstructure acting as a trigger for the ply degradation.

Understanding the crack initiation and propagation concepts requires a solid basis of fracture mechanics. Unfortunately, a complete and rigorous explanation of the physics and the mathematical tools involved in the fracture process is out of the scope of this document. Nevertheless, some basic ideas, such as the concept of quasi-brittle material, and the different manners of modeling damage used in computational micromechanics are briefly explained along the next paragraphs.

About quasi-brittle materials

Perhaps, a good starting point is the concept of *fracture process zone* (FPZ), developed by Irwin (1948), among others, and extended by Hillerborg et al. (1976), who generalized the work of Dugdale (1960) and Barenblatt (1962) trying to understand the fracture behavior of materials like concrete and rocks. Similar to the model of Dugdale-Barenblatt, Hilleborg proposed the existence of a thin region ahead of the crack tip, the FPZ, where the material shows a progressive softening according to increasing strains due to damage (usually microcracks). The extension of the FPZ depends on the material microstructure and nature and was defined by Hilleborg according to a characteristic length,

$$l_{\text{FPZ}} = \frac{EG_c}{\sigma_t^2} \quad (2.26)$$

, where E , σ_t and G_c are, respectively, the material elastic modulus, tensile strength and fracture energy. The relative size of the FPZ respect to the plastic zone and to the structure size will determine the fracture behavior. In the case of brittle materials, such as ceramics or glass, both the FPZ and the plastic region are very small and, hence, small strain increments lead to the quick coalescence of the microcracks into a macroscopic one that grows. In this case, Linear Elastic Fracture Mechanics (LEFM) provides a good description of the fracture process. In the case of ductile materials, such as metals, the fracture process zone is small too, nevertheless the extension of the plastic region, very often showing strain-hardening, surrounding the crack tip is relevant. Depending on the extension of the

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Figure 2.13: Types of fracture according to the FPZ relative dimensions. From left to right, brittle, ductile and quasibrittle material. Schematic of stress distribution along the crack line is shown at the bottom. Adapted from [Bazant and Planas \(1998\)](#).

plastic zone, small-scale yielding (SSY) techniques or full Elasto-Plastic Fracture Mechanics (EPFM) techniques, such as the J-integral CTOD approach, must be used ([Anderson, 2005](#)).

However, there is a third type of behavior observed in many materials such as concrete, rocks or epoxy resins, which are known as quasi-brittle materials ([Bazant and Planas, 1998](#)). Unlike metals, where the FPZ is quite small compared to the plastic region, in quasi-brittle materials the FPZ represents almost the whole non-linear region. In addition, the size of the non-linear region, almost equivalent to the FPZ, in quasi-brittle materials is quite large compared to metals and very often it comprises the whole cross section of the structure (Figure 2.13).

Cohesive vs smeared crack

Trying to understand the fracture behavior of quasi-brittle materials, Hilleborg redefined the Dugdale-Barenblatt cohesive crack model (he called it fictitious crack model) in terms of normal stresses that are transferred between the crack sides according to a stress-displacement law $\sigma_N = f(\delta)$. Hilleborg modeled the normal stress σ_N using a monotonically decreasing function according to the crack opening δ , where the area under this curve corresponds to the material fracture energy, G_C . Therefore, in the cohesive crack model, the stress is not assumed to fall to zero right after reaching the value of the material strength. After cracking, the stress at the crack is calculated by the fracture energy curve $\sigma_N = f(\delta)$.

Figure 2.14: Schematic of different manners used to approach the behavior of a real crack. (a) Cohesive crack (strong discontinuity) and (b) Smeared crack (continuum damage).

Although Hilleborg stated that the crack may grow anywhere, even if no initial macrocrack is present, it is worth to mention that when Hilleborg model is implemented in a finite element code, after cracking, a node must be split in two and therefore a strong discontinuity is introduced in the model, which means that the crack shows two different sides making the stress transfer unfeasible (Figure 2.14a). If the direction of crack is not known in advance, the direction in which the energy release rate is maximum should be calculated and if it does not propagate through nodes, adaptive remeshing will be required at each step.

Almost at the same time, [Bazant and Oh \(1983\)](#) proposed an alternative approach to the fracture process, the crack band model, where the crack is represented as a large set of microcracks of infinitely small opening that are continuously distributed or smeared over a band of a certain width placed ahead of the crack tip. Therefore, instead of tracking individual macrocracks, the crack evolution is modeled as a progressive degradation of the material stiffness (strain softening) according to increasing strains (Figure 2.14b).

Unlike the cohesive crack model, where the traction-displacement law that controls the crack opening can be directly related to the fracture energy, G_C , which is a material property, the crack band model uses a stress-strain law and, hence, it requires the usage of a volumetric fracture energy, g_C , which is not a material property. Therefore, when the model is programmed in a finite element code, the damage is propagated in a single row of elements whose size will control the amount of energy that is dissipated. This mesh dependency is avoided introducing a regularization parameter, a characteristic length, l_{ch} , associated with an integration point, which is

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based on the element geometry and formulation (i.e. in ABAQUS typical length of a line across an element for a first-order element and half of the same typical length for a second-order element).

$$g_C = \frac{G_C}{l_{ch}} = \int \sigma(\epsilon) d\epsilon \quad (2.27)$$

The characteristic element length has an upper limit defined by the material properties in order to avoid physically incorrect and numerically ill-posed models. Therefore,

$$l_{ch} \leq \frac{2EG_C}{\sigma_{yt}^2} \quad (2.28)$$

This two strategies are used in this work to model two microconstituents, the polymer matrix and the fiber/matrix interface. The former uses a continuum isotropic damage coupled with plasticity and relying in the smeared crack concepts, whereas the latter takes advantage of the Hilleborg's cohesive crack approach. A general explanation of both is given along the next paragraphs.

2.3.3 Modeling coupled damage and plasticity

Polymers, like other quasi-brittle materials, exhibit a different mechanical behavior whether they are loaded in tension or compression. Although the difference between compression strength and tension strength for a common epoxy is not as large as in the case of other quasi-brittle materials, such as concrete, the amount of plastic strain and hence the failure mechanisms are completely different.

Under pure tension, the polymer behaves as an elastic solid until the tension strength value is reached. This point, corresponds to the onset of material degradation, this time in the form of microcracking. Increasing the displacement lead to continuous nucleation and coalescence of microcracks producing a macroscopic softening of the material. However, under pure compression, there is a plastic hardening after the yield limit followed by a softening and the final material failure. The fracture process is dominated by the evolution of shear bands where the plastic strain is concentrated up to the final failure of the polymer, normally in a localized plastic band where the material suffers severe crushing. The amount of plastic strain before failure(crushing) can easily reach values larger than 35% (Figure 2.15).

Among the different constitutive equations available in the literature (Mohr-Coulomb, Drucker-Prager, etc), the damaged-plasticity model includes a number of characteristics that make it specially suited for modeling quasi-brittle materials, such as epoxy polymers. The damage-plasticity model, first employed for quasibrittle materials by Lubliner et al. (1989), is a modification of the pressure dependent Drucker-Prager plasticity yield criterion Drucker and Prager (1952) and, hence, it allows the definition of different tension and compression behavior. However, it uses concepts of isotropic (scalar) damaged elasticity in combination with isotropic tensile and compressive plasticity to represent the inelastic behavior of the quasibrittle material, including the irreversible process that occurs during fracturing. This continuum, plasticity-based, damage model assumes that, at low confining pressure, the main two irreversible processes affecting the material are tensile cracking (actually coalescence of microcracking) and shear yielding (ending with compressive crushing).

Material degradation and uniaxial behavior

Under uniaxial conditions, the evolution of the yield surface is controlled by two hardening variables, the tensile and compressive equivalent plastic strains, $\bar{\epsilon}_t^{\text{pl}}$ and $\bar{\epsilon}_c^{\text{pl}}$, respectively. The uniaxial behavior of the damage-plasticity model is illustrated in Figure 5.3, including the material degradation.

Under pure tension, the material behaves as an elastic solid until the tension strength value is reached. This point, corresponds to the onset of material degradation, this time in the form of microcracking. Increasing the displacement lead to continuous nucleation and coalescence of microcracks producing a macroscopic softening of the material that can be observed in the σ_t decay (Figure 2.15a).

Under pure compression, the stress-strain is again linear until the compression strength value is reached. However, in this case the material is able to undergo plastic deformation. Typically, material experiences some hardening until the ultimate compression stress is reached. This point corresponds to the onset of compression damage (crushing). The material collapse is modeled by means of a σ_c decay (Figure 2.15b).

In both cases, together with the σ_t and σ_c decays, two independent uniaxial damage laws, $d_t = d_t(\bar{\epsilon}_t^{\text{pl}})$ and $d_c = d_c(\bar{\epsilon}_c^{\text{pl}})$ must be specified. Therefore, effective uniaxial cohesion stresses, $\bar{\sigma}_t$ and $\bar{\sigma}_c$, under uniaxial tension and compression loading,

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Figure 2.15: Schematic of the uniaxial tension (a) and compression (b) stress-strain response normally used in the damage-plasticity model. Regions where damage is activated are highlighted in green.

determining the size of the yield (or failure) surface, are:

$$\bar{\sigma}_t = \frac{\sigma_t}{1 - d_t} = E_0(\epsilon_t - \bar{\epsilon}_t^{pl}) \quad (2.29a)$$

$$\bar{\sigma}_c = \frac{\sigma_c}{1 - d_c} = E_0(\epsilon_c - \bar{\epsilon}_c^{pl}) \quad (2.29b)$$

It is worth to mention that, within the context of the scalar-damage theory, the stiffness degradation is isotropic and characterized by a single degradation variable, d . In the absence of damage $d = 0$ whereas a fully damaged state means that $d = 1$. The evolution of d is controlled by the equivalent plastic strain $\bar{\epsilon}^{pl}$ and the effective

stress $\bar{\sigma}$ defined as:

$$\bar{\epsilon}^{\text{pl}} = [\bar{\epsilon}_t^{\text{pl}}, \bar{\epsilon}_c^{\text{pl}}]^T \quad (2.30a)$$

$$\bar{\sigma} = \mathbf{D}_0^{\text{el}} : (\epsilon - \bar{\epsilon}^{\text{pl}}) \quad (2.30b)$$

, where $\mathbf{D}_0^{\text{el}}$ is the undamaged elastic stiffness tensor and ϵ and ϵ^{pl} are the total and plastic strain tensors, respectively. Therefore $d = d(\bar{\sigma}, \bar{\epsilon}^{\text{pl}})$. Of course, the Cauchy stress tensor σ is related to the effective stress tensor by means of the damage variable d according to:

$$\sigma = (1 - d)\bar{\sigma} \quad (2.31)$$

Multiaxial behavior and yield surface evolution

Regarding the general multiaxial stress conditions, since the definition of the scalar degradation variable d must be consistent with the uniaxial monotonic responses d_t and d_c , the model assumes that

$$(1 - d) = (1 - s_t d_c)(1 - s_c d_t) \quad (2.32)$$

, where $s_t = 1 - w_t r(\bar{\sigma})$ and $s_c = 1 - w_c(1 - r(\bar{\sigma}))$ are given in terms of the stiffness recovery factors w_t and w_c ($w_t = 0$ and $w_c = 1$ by default) and the stress weight factor ($0 < r(\bar{\sigma}) < 1$).

The damage-plasticity model yield function was proposed by [Lubliner et al. \(1989\)](#) and modified by [Lee and Fenves \(1998\)](#) to account for the different strength evolution under tension and compressive loads. In terms of the effective stresses and the equivalent plastic strains, the yield function can be expressed as

$$\Phi(\bar{\sigma}, \bar{\epsilon}^{\text{pl}}) = \frac{1}{1 - \alpha} \left(\sqrt{3J_2} + \alpha I_1 + B(\bar{\epsilon}^{\text{pl}}) \langle \bar{\sigma}_I \rangle - \gamma \langle -\bar{\sigma}_I \rangle \right) - \bar{\sigma}_{\text{yc}}(\bar{\epsilon}_c^{\text{pl}}) = 0 \quad (2.33)$$

, wherein I_1 stands for the first invariant of the equivalent stress tensor, J_2 is the second invariant of the equivalent deviatoric stress tensor, α is the pressure-sensitivity parameter of the original Drucker-Prager yield criterion ([Drucker and Prager, 1952](#)), $\bar{\sigma}_I$ is the algebraically maximum eigenvalue of $\bar{\sigma}$, $\langle \cdot \rangle$ the Macaulay brackets (returning the argument if positive and zero otherwise) and $B(\bar{\epsilon}^{\text{pl}})$ is a

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function of the effective tensile and compressive uniaxial yield stresses, $\bar{\sigma}_t$ and $\bar{\sigma}_c$, defined as

$$B(\bar{\epsilon}^{pl}) = \frac{\bar{\sigma}_{yc}(\bar{\epsilon}_c^{pl})}{\bar{\sigma}_{yt}(\bar{\epsilon}_t^{pl})}(1 - \alpha) - (1 + \alpha) \quad (2.34)$$

It is worth to mention that, under biaxial compression stress state, equation 2.33 reduces to the original Drucker-Prager yield condition [Drucker and Prager \(1952\)](#), wherein α can be expressed in terms of the polymer internal friction angle β according to $\tan(\beta) = 3\alpha$. The internal friction angle controls the hydrostatic pressure dependence of the plastic behavior of the material. Simultaneously, α can be related to the biaxial compression behavior according to

$$\alpha = \frac{\sigma_{yb0} - \sigma_{yc0}}{2\sigma_{yb0} - \sigma_{yc0}} \quad (2.35)$$

, where σ_{yb0} and σ_{yc0} are the initial biaxial and uniaxial compression strength, respectively.

Finally, the coefficient γ enters the yield function only for stress states of triaxial compression. This coefficient can be determined as a function of K_c , the ratio between the yield conditions $\rho(\Theta, K_c)$ along the tensile and compressive meridians (TM and CM, respectively), according to:

$$K_c = \frac{\gamma + 3}{2\gamma + 3} \quad (2.36)$$

The value of K_c affects the yield surface in the deviatoric plane only under triaxial compression stress states(Figure 2.16b). If $K_c = 1$, $\gamma = 0$ and the yield surface becomes independent of the lode angle Θ . In other words, the behavior of the damage plasticity model in the deviatoric plane corresponds to the Drucker-Prager criterion.

Regarding the potential flow rule, it can be formulated as

$$\dot{\epsilon}^{pl} = \dot{\lambda} \frac{\partial G(\bar{\sigma})}{\partial \bar{\sigma}} \quad (2.37)$$

, where $\dot{\lambda}$ is a non-negative plastic multiplier and G is a flow potential defined as

(a)

(b)

Figure 2.16: a) Initial yield surface under plane stress ($\sigma_3 = 0$), b) Yield surfaces under triaxial compression stress state in the deviatoric plane (π -plane), corresponding to different values of K_c .

2.3. Constitutive behavior of FRPs constituents

$$G = \sqrt{(\epsilon \sigma_{yt0} \tan(\psi))^2 + 3J_2^2} + \frac{I_1}{3} \tan(\psi) \quad (2.38)$$

, where σ_{yt0} is the tensile strength, ψ is the dilation angle and ϵ is the eccentricity parameter, that defines the rate at which the flow potential tends to a straight line (linear Drucker-Prager). This hyperbolic yield criterion is a continuous combination of the maximum tensile stress condition of Rankine (tensile cutoff) and the linear Drucker-Prager condition at high confining stress. Therefore, at the meridian plane, the yield surface intersects the pressure axis at 90° , similar to the Rankine criterion. This flow potential G is continuous and smooth, providing a unique flow direction. The damage-plasticity model assumes non-associated plasticity in the meridional plane, however, associated flow can be retrieved setting the dilation angle ψ equal to the internal friction angle ($\psi = \beta$). More information can be found in [Lublinter et al. \(1989\)](#) and [Lee and Fenves \(1998\)](#).

2.3.4 Modeling fiber/matrix interface

Despite of the advantages of the smeared crack models, the cohesive fracture models are very suitable to simulate the progressive interface fracture upon loading at the interface between dissimilar materials, since the crack path is clearly defined. The cohesive crack can be implemented in finite elements codes by means of different methods like extended finite elements, cohesive elements, strong embedded discontinuities, etc. However, regarding the fiber/matrix interfaces, the most common way to simulate the crack initiation and propagation is based on cohesive elements. Based on Hilleborg's model, the mechanical response of this cohesive elements is formulated in terms of a stress-displacement law. The cohesive element behaves as an elastic solid upon damage initiation, which can be activated by several failure criteria (maximum stress, maximum strain, quadratic stress, quadratic strain, etc). Once the damage begins, the cohesive element stiffness is reduced according to a damage scalar parameter, d , that ranges between 0 (absence of damage) and 1 (element fully damaged).

In the case of several failure modes take place at once, the mixed mode damage evolution law is required and the damage parameter is calculated according to an effective displacement, $\bar{\delta}$ defined as the norm of the displacement jump vector across the interface as

$$\bar{\delta} = \sqrt{\langle \delta_N \rangle^2 + \delta_S^2 + \delta_T^2} \quad (2.39)$$

where $\langle \bullet \rangle$ stands for the Macaulay brackets indicating that the displacement jump in the normal direction is only taken into account when it leads to crack separation. In other words, normal compression loading has no effect on the interface damage propagation.

The evolution of the damage variable is controlled by the fracture energy associated with the cohesive material. In particular, assuming a linear softening $\bar{\tau}(\bar{\delta})$ law (Figure), the maximum displacement jump across the interface, $\bar{\delta}_f$, required to produce the final failure of the element can be estimated as

$$\bar{\delta}_f = \frac{2\bar{G}_C}{\bar{\tau}} \quad (2.40)$$

where G_C and $\bar{\tau}$ are the effective fracture energy and stress, respectively. The effective stress is the norm of the stress vector at the interface, whereas the effective fracture energy G_C is calculated using the mixed mode law,

$$G_C = f(G_{IC}, G_{IIC}, G_{IIIC}, \eta) \quad (2.41)$$

where G_{IC} , G_{IIC} and G_{IIIC} are the fracture energy of the different individual failure modes and η is a fitting parameter.

For composite materials, the effective fracture energy G_C is often calculated using the [Benzeggagh and Kenane \(1996\)](#) model (Equation 2.42), which is particularly suited when the critical fracture energies during deformation purely along the first and the second shear directions are the same, $G_{IIC} = G_{IIIC}$ (Figure).

$$G_C = G_{IC} + (G_{IIC} - G_{IC}) \left(\frac{m^2}{1 + 2m^2 - 2m} \right)^\eta \quad (2.42)$$

, where $m = \langle \delta_N \rangle / \sqrt{\delta_S^2 + \delta_T^2}$ represents the mixed-mode ratio, quantifying the proportion of mode I to mode II, and η is a fitting parameter, that ranges between $1.5 < \eta < 2.3$ depending on the ductility of the matrix.

Under the assumption of the linear softening, the damage can be estimated as

$$d = \frac{\bar{\delta}_f(\bar{\delta} - \bar{\delta}_0)}{\bar{\delta}(\bar{\delta}_f - \bar{\delta}_0)} \quad (2.43)$$

where $\bar{\delta}_0$ represents the effective displacement at the onset of damage.

Figure 2.17: Schematic of the linear damage evolution represented by the cohesive interface

Figure 2.18: Schematic of the [Benzeggagh and Kenane \(1996\)](#) mixed mode law for damage evolution represented by the cohesive interface

2.3.5 Modeling water absorption in FRPs

The water absorption can affect the mechanical response of the FRP in several ways. In the case of GFRP or CFRP, where the fibers are assumed to be insensitive to water uptake, the moisture effect will appear through the polymer matrix and fiber/matrix interface.

In the case of the polymer matrix, the water produce two main problems, plasticization and swelling. The former, is reflected in a drop of the glass transition temperature T_g^{WET} . In polymers, the glass transition temperature T_g is related to the chain flexibility and the amount of free volume into the material. By absorbing a miscible liquid whose free volume is larger than the pure polymer (such as water), T_g will be lowered (Kelley and Bueche, 1961).

At fixed strain rate, when temperature is a few degrees below T_g the plastic flow in glassy polymers can be approximated as

$$\sigma_y \approx \frac{\sigma_0}{T_g}(T_g - T) \quad (2.44)$$

which explains the experimental results given by several authors on different thermoplastic and thermoset polymers (citation to Bauwens1972, Argon and Bessonov1977 and Matsuoka1981).

In the case of the computational micromechanical models based on finite elements, this effect is introduced by modifying the Young modulus E and the polymer yield stress, σ_y according to temperature and is specially relevant if the composite service temperature is close to this new T_g^{WET} since the mechanical properties will be reduced considerably.

On the other hand, the swelling of the matrix is considered using the coefficient of moisture expansion (CME) in a similar manner to the thermal expansion that is included by using the coefficient of thermal expansion (CTE). The increment of the moisture content (the water absorption) ΔM is translated in terms of principal strains according to $\epsilon_i = \beta_i \Delta M$.

Back to the computational micromechanical models based on finite elements, the mismatch between the polymer and fibers CME will result in another source of residual stresses, together with the thermal stresses. Assuming that both residual thermal and swelling stresses produce small deformations and the polymer remains

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linear elastic, the superposition principle can be applied in order to compute the total residual strain,

$$\epsilon_i = \epsilon_i^T + \epsilon^M = \alpha_i \Delta T + \beta_i \Delta M \quad (2.45)$$

At this point, the effective coefficients of thermal expansion, α_i^* are introduced. This coefficients produce total strains in one temperature drop, that are equivalent to the effect of moisture and temperature acting together. Therefore, Equation 2.45 can be rewritten as

$$\alpha_i^* \Delta T = \alpha_i \Delta T + \beta_i \Delta M \quad (2.46)$$

Polymers are often considered as isotropic materials ($\alpha_1 = \alpha_2 = \alpha_3$ and $\beta_1 = \beta_2 = \beta_3$) and, hence, both the thermal expansion and the swelling are independent of the material direction. Therefore, the matrix effective coefficients of thermal expansion can be defined as,

$$\alpha_m^* = \frac{\alpha \Delta T + \beta \Delta M}{\Delta T} \quad (2.47)$$

Since fibers are considered to absorb no moisture, only their thermal expansion coefficients, $\alpha_{f,1}$, $\alpha_{f,2}$ and $\alpha_{f,3}$ are considered. Therefore, the homogenized effective thermal expansion coefficients of the composite can be obtained, using a simple rule of mixtures, according to Equation .

$$\alpha_1^* = \alpha_{f,1} V_f + \alpha_m^* (1 - V_f) \quad (2.48a)$$

$$\alpha_2^* = \alpha_{f,2} V_f + \alpha_m^* (1 - V_f) \quad (2.48b)$$

$$\alpha_3^* = \alpha_{f,3} V_f + \alpha_m^* (1 - V_f) \quad (2.48c)$$

So far, the general state-of-the-art of computational micromechanics based on Finite Elements simulations have been exposed. Both the RVE generation procedures and the constitutive models of the FRP constituents have been described. In the next chapters, both the experimental techniques used to characterize the micro-constituents and the numerical models required for the different failure modes are explained.

Material and experimental techniques

The aim of this chapter is to explain the basis of the experimental techniques used in this work to obtain the mechanical response of the composite constituents. After introducing the AS4/8552 composite material, the experimental techniques are presented, divided in two groups, *in-situ* and *ex-situ*. The in-situ techniques stand for characterization processes allowing the experiments to be directly carried out on the composite cross section and, hence, they have a pure micromechanical nature. They are the instrumented nanoindentation, the fiber push-in and the micropillar compression tests and their study and application to the AS4/8552 are one of the main objectives of this work.

On the other hand, two additional testing techniques are used in this work, the single fiber tensile test and the neat resin compression. Whereas the former is used to obtain the fiber properties related to the tensile behavior, namely the longitudinal tensile modulus and strength and the mode I fracture toughness, the latter is used to check the results obtained by some of the in-situ techniques.

3.1 Material

The material studied in this work is carbon/epoxy unidirectional laminate of aeronautical grade made of AS4/8552 pre-impregnated (pre-preg) laminae. The nominal fiber volume fraction is 58% and the nominal cured ply thickness is 0.178 mm. The laminate was prepared by manually stacking 12 plies using a unidirectional sequence $[0]_{12}$. The laminate was cured following the supplier recommendations specified in the material data sheet, 2 hours at 130°C followed by 2 hours at 180°C with 6.2

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bars of pressure. Since this well-known pre-impregnated material system has been widely used in the aeronautical industry and subject of research most of its ply properties are directly provided by the manufacturer or can be easily found in the literature [Marlett et al. \(2011\)](#); [Lopes et al. \(2015\)](#), providing a good reference for the micromechanical predictions.

Regarding the microconstituents, the 8552 epoxy resin is a blend of a high functionality epoxy resin TGMDA (Tetraglycidyl methylenedianiline) and a lower functionality epoxy resin TGpAP (Triglycidyl p-aminofenol). Two different amines, the 4,4'-diaminodiphenylsulfone (DDS) and 3,3'-diaminodiphenylsulfone are used as curing agents. In addition, the 8552 epoxy resin has been modified by the supplier with the incorporation of thermoplastic particles, among other additives. As a result, the 8552 is a glassy polymer that offers outstanding mechanical properties, including high stiffness and enhanced fracture toughness and impact performance.

On the other hand, the AS4 carbon fiber, manufactured by Hexcel, is a high-strength, high strain intermediate modulus continuous fiber obtained from polyacrylonitrile (PAN) precursor. The AS4 carbon fiber is usually surface treated and sized to improve its interlaminar shear properties, handling characteristics, and structural properties.

3.2 *In-situ* micromechanical experimental techniques

The accuracy of the determination of ply properties in unidirectional composites by means of computational micromechanics relies on the actual properties of the fibre, matrix and fibre/matrix interface. In a first approximation, they could be obtained by means of tests carried out in either fibers or matrix specimens or in specific coupons for the interface. For instance, tensile tests on individual fibre filaments or yarns can be used to determine the elastic modulus and strength of the fibre, and neat resin specimens can be used to measure the stiffness and the strength in tension, compression and shear as well as the fracture toughness of the matrix. In the case of the fibre/matrix interface properties, manufacturing of additional *ad hoc* specimens is often required. This is the case of a single fibre embedded in a resin coupon for the pull-out test ([Désarmot and Favre, 1991](#)) or the resin drop cured on a single fibre for the microbond test ([Miller et al., 1987](#)). In both cases, sample preparation is not straightforward and very often is not feasible, since the liquid resin is hardly ever provided by the supplier.

In addition, matrix properties extracted from neat resin specimens or interface properties obtained from pull-out or microbond tests could not be representative of the actual properties within the unidirectional ply because of the differences in the manufacturing process, the curing conditions and the residual stresses. For instance, the matrix and interface properties depend on the local thermal history during curing that could be modified due to the large fibre volume fraction in the ply.

In view of this, the optimum solution relies on the use of nanomechanical testing techniques to measure the *in situ* matrix, fibers and interface properties within the actual composite ply. However, these type of tests are not exempt of difficulties, requiring sometimes additional macroscopic testing in order to provide a full description of the elasto-plastic behavior of the material (Rodríguez et al., 2012b). Along the next paragraphs the several micromechanical techniques, the instrumented nanoindentation, micropillar compression and fiber push-in are explained. These techniques have been used and sometimes combined in this work to obtain the properties of the constituents.

3.2.1 Matrix characterization by instrumented nanoindentation

In this technique, an spherical, conical or pyramidal rigid indenter is used to penetrate the surface of the material. Measuring the applied load P and the contact area A_c , the material hardness H can be obtained according to Equation 3.1 (Tabor (1970)). However, the problem now is how to determine the real contact area.

$$H = \frac{P}{A_c} \quad (3.1)$$

Concerned by this problem and taking advantage of the modern nanoindenters, which allow the continuous measurement of indentation load P and penetration depth, h , Oliver and Pharr Oliver and Pharr (1992), developed a method where the contact area A_c is obtained using an indentation load-displacement curve as the one shown in Figure 3.1. To this end, the maximum indentation load $P_{\max}$, the maximum penetration depth $h_{\max}$ and the stiffness of the unloading curve S are required. Oliver and Pharr proposed to fit the unloading curve using a non-linear relationship, where $P = \alpha(h - h_f)^m$ and the parameters α , h_f and m are determined using a least-squares fitting. The stiffness $S = dP/dh$ is obtained calculating the derivative at the maximum load. Once the stiffness is known, the contact depth is

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determined as $h_c = h_{\max} - \epsilon \frac{P_{\max}}{S}$, where ϵ depends on the tip geometry used for the indentation. Since the geometry of the tip is known, the relationship between the contact area A_c and the contact depth h_c is known as well and the material hardness can be obtained. On the other hand, the reduced elastic modulus is calculated based on Sneddon's contact law according to Equation 3.2.

$$E^* = \frac{S\sqrt{\pi}}{2\sqrt{A_c}} \quad (3.2)$$

Finally, the material Young modulus is obtained through Equation 3.3, that takes into account the deformation of the indenter, whose Young modulus and Poisson ratio are E_i and ν_i respectively.

$$\frac{1}{E^*} = \frac{1 - \nu^2}{E} + \frac{1 - \nu_i^2}{E_i} \quad (3.3)$$

Figure 3.1: (a) Typical indentation load-displacement curve and (b) scheme of Berkovich indentation showing pile-up/sink-in behavior affecting the contact depth (h_c) and hence the contact area of indentation. Adapted from Rodríguez et al. [Rodríguez et al. \(2012b\)](#).

Although the Oliver and Pharr method has been widely used in material science, it requires special attention if it is applied to frictional materials such as bulk metallic glasses, or polymers, where the pile-up/sink-in effects can take place (Figure 3.1b). Based on this, in this section the work developed by [Molina-Aldareguía et al. \(2011\)](#) and [Rodríguez et al. \(2012b\)](#), among others, is summarized, underlying the limitations of instrumented indentation alone to determine the full constitutive behavior of epoxy polymers, which constitutes the motivation of this work.

Epoxies, as other cohesive frictional materials, are brittle under tension loads while, under compression, they exhibit a marked plastic behavior that leads to strain localization in shear bands. Since the yield surface is affected by the hydrostatic stress, its evolution can be modeled using a Drucker-Prager yield criterion, which means that, assuming isotropic elasto-plastic behavior, four parameters are required to describe the constitutive behavior of the polymer: while the Young modulus (E) and the Poisson's ratio (ν) describe the elastic behavior, the cohesion (d) and the internal friction angle (β) determine the plastic yield surface.

The problem of obtaining the elasto-plastic properties of cohesive-frictional materials (such as polymers, bulk metallic and ceramic glasses) from nanoindentation, was analyzed by [Rodríguez et al. \(2012b\)](#) who performed a set of parametric studies using the Finite Element (FE) method to understand the pile-up/sink-in phenomena. The study concluded that, the pile-up/sink-in will depend on the tip geometry (pyramidal, conical spherical, etc.), the plasticity index (σ_{yc}/E^*) and the internal friction angle (β), which means that the analysis of the indentation load-displacement curves alone (as shown in Figure 3.1a) cannot provide, independently, all the information required to determine the other three parameters that describe the elasto-plastic behavior. This is partially due to the uncertainty in the pile-up/sink-in behavior that can take place in the material surrounding the indenter ([Cheng and Cheng \(1998\)](#)), as schematically illustrated in (Figure 3.1b). As a result, the actual contact area between the tip and the material, A_c , is not known and specific assumptions should be formulated to determine the modulus and hardness of the material.

In order to avoid the uncertainty introduced by the pile-up/sink-in behavior, [Rodríguez et al. \(2012b\)](#) proposed the use of two parameters that can be easily determined from the indentation curve: the apparent hardness H_{ap} and the ratio of the elastic and total work of indentation W_e/W_t .

The real hardness H is given by Equation 3.1, where A_c stands for the real contact area that is affected by the pile-up/sink-in behavior. The apparent hardness H_{ap} is given by Equation 3.4, where A_{max} is the projected area of the indenter at the maximum indentation depth h_{max} . Additionally, W_e/W_t can be easily determined by integration of the areas under the load-displacement curve (Figure 3.1a).

$$H_{ap} = \frac{P_{max}}{A_{max}} \quad (3.4)$$

It should be noted that the ratio between H and H_{ap} can be directly related to

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the pile-up/sink-in parameter c_p through Equation 3.5:

$$H = \frac{H_{ap}}{c_p^2} \quad (3.5)$$

where c_p measures the ratio between the square roots of the real and apparent contact areas through Equation 3.6:

$$c_p = \sqrt{\frac{A_c}{A_{max}}} \quad (3.6)$$

Based on these two parameters, [Rodríguez et al. \(2012b\)](#) proposed a set of master curves (Figure 3.2) relating E , σ_{yc} and β , obtained by means of a parametric study of the indentation process using FEM.

The master curves in Figure 3.2 demonstrate the impossibility of determining the elasto-plastic response of cohesive frictional materials from instrumented indentation, due to the uncertainty in the pile-up parameter c_p . This is because the *Oliver and Pharr* method, [Oliver and Pharr \(1992\)](#), only provides a good estimate of the contact area if $W_e/W_p > 0.5$, as shown in Figure 3.2a. Additionally, the ratio between hardness H and the uniaxial compressive yield stress σ_{yc} is not unique, as it depends on the internal frictional angle β as shown in Figure 3.2c. This limitation could be potentially overcome by measuring the real contact area A_c by, for instance, atomic force microscopy, but viscoelastic effects prevent this approach in polymers, as substantial recovery of the indentation imprint can take place with time.

Thus, only in those cases where one of the parameters, E , σ_{yc} or β , is known, the rest can be unequivocally determined using the corresponding master curve (Figure 3.2). For instance, if E is known, the contact area A_c and hence, the pile up parameter c_p , could be determined from the unloading stiffness S (Figure 1) through Equation 3.2:

However, for epoxy resins, for which W_e/W_t is typically > 0.5 , the dependency of c_p with β is weak (see Figure 3.2a) and this approach becomes unfeasible. Therefore, in conclusion, either σ_{yc} or β must be known, in order to obtain the other parameter from nanoindentation, using Figure 3.2b.

One way of tackling this problem is macroscopic testing [Rodríguez et al. \(2012a\)](#) of resin coupons. However, this approach requires the manufacturing of *ad-hoc* resin coupons and one cannot ensure that the properties are the same as those of the resin inside the composite material, since the local curing conditions might differ.

Figure 3.2: Evolution of (a) pile up parameter and (b) apparent hardness constraint factor (H_{ap}/σ_{yc}) as a function of W_e/W_t for different internal friction angle β . (c) Evolution of the constraint factor (H/σ_{yc}) as a function of the plasticity index (σ_{yc}/E^*) for different values of β . reprinted with permission from Rodríguez et al. (2012b).

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However, sample preparation is often not feasible, since the liquid resin is hardly ever provided by the supplier.

In view of this, in this work, we propose combining nanoindentation with resin micropillar compression tests in order to determine with high accuracy the polymer elasto-plastic behavior.

3.2.2 Micropillar compression test

The main idea under the micropillar compression tests is quite simple, performing uniaxial compression tests on samples having micron-size dimensions. However, the difficulties associated to sample manufacturing delayed the application of this type of micromechanical tests until 2004, [Uchic and Dimiduk \(2005\)](#), showing the potential of this technique in order to study the mechanical properties at the micro scale. Since then, micropillar compression tests have become very popular not only to study the effect of material orientation and grain size in the mechanical properties of a single crystal but also to study plasticity in brittle materials without the need of using complex tests. However, so far, most of the micropillar compression tests reported in the literature have been carried out in hard materials, such as metals and ceramics, that can withstand large stresses. In this work the usage of micropillar compression tests to study the mechanical response of a soft material, an epoxy resin, is investigated.

Regarding the micropillar manufacturing, different techniques such as lithography or directional solidification can be used. However, since the idea of this work is to develop a micromechanical testing methodology, *in-situ* in the composite material, a Focused Ion Beam (FIB) is used to carve the micropillars on the resin rich areas of the composite cross-section. This technology allows direct micro and nano-scale deposition or materials removal anywhere on a solid surface.

The FIB produces and directs a stream of high-energy ionized atoms of a relatively massive element, normally Ga^+ , that are accelerated in the range of 0.5 to 50 keV using electric fields. The ions hit the material surface sputtering away the atoms of the target material in what is called the milling process. The ion greater mass allows to easily expel surface atoms from their positions and produces secondary electrons from the surface, allowing the ion beam to image the sample before, during, and after the process. The typical resolution of modern FIB is in the range of 5 nm.

Modern FIB are normally equipped with a dual beam system, where a very precise FIB with etching and deposition capabilities is combined with a ultra-high resolution field emission Scanning Electron Microscopy (SEM). Normally the SEM beam is placed in the vertical direction, whereas the ion beam is placed at 52° respect to it (Figure 3.3). The dual beam system allows simultaneous imaging and milling. The samples are placed in vacuum chamber that guarantees ultra-high vacuum operating conditions allowing the usage of different detectors and imaging techniques such as electron backscatter diffraction (EBSD), scanning transmission electron microscope (STEM), etc. Finally, a rotating stage permits to tilt the sample at different angles in order to adapt the milling and imaging process according to the user's requirements (sample geometry, detection mode, etc.).

Figure 3.3: Schematic diagram of the FIB/SEM dual beam apparatus. The electron and the ion columns are placed at an angle of 52° . The sample is mounted on a rotating holder that can be tilted, rotated and translated

Pillar manufacturing

The pillars can be carved using annular or lathe milling. In the former, circular ring patterns perpendicular to the ion beam are used during the process 3.4. The samples does not move during the milling, remaining perpendicular to the ion beam,

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which sweeps the surface in a circular movement. The annular groove becomes deeper and deeper at each pass of the ion beam. The diameter of the annular pattern is reduced progressively and the ion current is adapted to control the depth until the desired pillar geometry is obtained. In the lathe milling, Figure 3.5, a large diameter preform ring of a depth approximating the targeted final pillar height is fabricated using a circular milling pattern. Then, the samples is tilted to a small angle relative to the ion beam and a series of small rectangular cuts and rotations are used to remove material from the column's sidewalls in order to achieve the final desired geometry. After each rotation, the stage needs to be re-positioned for the next milling step.

The lathe milling allows an optimum control of the diameter and height of the pillar and is not sensitive to differences in milling rate, making it particularly advantageous for heterogeneous materials. However, the process is very time consuming (large pillars in hard materials can take more than 24 hours) and, in addition, since imaging the sample is required after each rotation step to re-position the stage, the sample will receive a large dose of ions that can modify the material structure.

On the other hand, the annular milling, which is not as precise as the lathe milling, produces slightly tapered pillars that can complicate the stress-strain analysis due to the non-uniform cross-sectional area. Other issues include rounding at the edges, different milling rates for heterogeneous materials, a non-uniform foot geometry complicating the height assessment, and difficulties in controlling the column height. However, the process is much faster than the lathe milling method and the ion dose can be significantly reduced since no imaging with the ion beam is necessary. For this reason, this is the preferred technique in most of the micropillar compression tests. In this work, all the pillars have been carved using the annular milling technique.

Figure 3.4: Schematic diagram of the FIB/SEM annular milling process

Figure 3.5: Schematic diagram of the FIB/SEM lathe milling process

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Pillar compression

Once the pillars are produced, they are placed in the nanoindenter in order to be compressed, normally using a flat punch tip (Figure 3.6). The nanoindenter records the load P and the displacement u simultaneously and, since the geometry of the pillar is measured using the SEM, the true stress-strain curve can be retrieved according to:

$$\epsilon = \ln(1 - u_p/H_0) \quad (3.7a)$$

$$\sigma = \frac{4P}{\pi D_0^2(1 - u_p/H_0)} \quad (3.7b)$$

where, D_0 and H_0 correspond to the initial top diameter and height of the pillar, respectively.

It is important to mention that u_p , the displacement of the pillar, does not coincide with u , the displacement measured by the nanoindenter, because u includes the displacement of the substrate u_s that acts as an elastic foundation. Therefore, $u_p = u - u_s$ where u_s can be estimated using Equation 3.8, proposed by Sneddon (Fei et al., 2012), where E_s and ν_s stand for the substrate's Young modulus and Poisson ratio, respectively, and A_p is the pillar cross section area.

The idea here is to assume that the pillar is acting on the substrate as it was an indenter, which means that the pillar is assumed to be completely rigid, resulting in a slight overestimation of the compliance. In addition, when the pillars are carved on the composite cross section, the substrate and the pillar are both made from the same material. This particularity together with other important effects such as the taper angle, the misalignment between the indenter and the pillar and the FIB-induced damage will be discussed in detail in the next chapter.

$$u_s = C_{\text{Sneddon}} P = \frac{\sqrt{\pi}(1 - \nu_s^2)}{2E_s\sqrt{A_p}} P \quad (3.8)$$

3.2.3 Interface characterization by instrumented nanoindentation

While a lot research has been carried out to improve the mechanical properties of carbon and glass fibers and of thermoset and thermoplastic matrices, optimization

Figure 3.6: Schematic diagram of micropillar compression test

of interface properties was more elusive because of the difficulties associated with the mechanical characterization. Nevertheless, it is worth to mention that the ratio between the interface surface and the material volume is $r = 2V_f/r_f$, being V_f the fiber volume fraction and r_f the fiber radius. Therefore, in a typical aeronautical grade composite, fiber diameter $d_f = 7\mu\text{m}$ and fiber volume fraction $V_f = 60\%$, $r = 1.71 \times 10^5 \text{ m}^2/\text{m}^3$. This means that there is more or less the surface of 25 professional football fields in one cubic meter of composite material. In view of this, the probability of damage triggered by interface fracture is at least remarkable and, hence, the development of sound experimental methods to measure the fiber-matrix interface properties appears an important task to design composites with improved performance.

Interface response of FRPs can be obtained using samples extracted from the real composite material or manufacturing additional *ad hoc* specimens. So far, different testing techniques have been used to determine the mechanical properties of fiber-matrix interfaces. They include the pull-out test [Kharrat et al. \(1997\)](#), the microbond test [Nishikawa et al. \(2008\)](#) and the fragmentation test [Zhou et al. \(2001\)](#). Nevertheless, the interface strength obtained from different tests often differ and even the results from one type of test are subjected to a large scatter [Pitkethly et al. \(1993\)](#), very likely because the analysis of the experimental data to obtain the interface properties is based on simplified analytical models. Moreover, it is not clear that tests based on coupons containing one fiber embedded in the matrix (pull-out, fragmentation or microbond) are representative of the actual mechanical and physico-chemical conditions induced by the local thermal history in real composites,

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since these *ad hoc* specimens do not capture the thermal stresses and the influence of the large fibre volume fraction in the ply.

To the knowledge of the author, the most suitable *in situ* techniques to measure the strength are the push-in and the push-out tests. Both techniques have been used in the past to measure the properties of composite materials (González et al., 2004) and, in particular, of FRPs (Rodríguez et al., 2012a; Canal et al., 2012b). Although both techniques are similar, they present as well some important differences in terms of sample preparation and results interpretation. Along the next paragraphs, both techniques are explained in order to justify their use.

Figure 3.7: Push-out test. Adapted from Canal (2011)

The push-out test

The push-out test requires the preparation of a thin slice perpendicular to the fibers with a thickness of the order of the fibre diameter. The sample is glued to a groovy holder in order to allow individual fibers to be pushed out from the composite cross section (Figure 3.7). The test is carried out using a nanoindenter, normally equipped with a flat tip, that monitors both the load and the displacement, applied to the fiber.

The shear strength of the interface can be obtained from the ideal load-displacement curve shown in Figure 3.8a. At the beginning of the test, the sample bends elastically until fiber compression starts. During this part a linear relationship between the load and the displacement is observed. Increasing the normal load until a critical value P_{crit} is reached, triggers the debonding and the load-displacement curve becomes nonlinear. At this moment, the shear stress drops to the residual friction stress in the debonded zone (red line in Figure 3.8b). Then, the region of maximum shear stress moves towards the bottom of the sample (blue line in Figure 3.8b). The nonlinear region finishes right after the moment of maximum load P_{max} , when the interface fracture reaches the bottom of the sample (Figure 3.8b). At this point, the whole interface is debonded. Increasing the displacement leads to some friction dissipation until the whole fiber is pushed out of the matrix. The interface strength T_d can be directly calculated using this P_{max} value and the interface area A_{int} as

$$T_d = \frac{P_{\text{max}}}{A_{\text{int}}} = \frac{P_{\text{max}}}{2\pi r_f t} \quad (3.9)$$

where r_f stands for the fiber radius and t is the thickness of the sample.

Although the procedure seems to be rather simple, with the exception of the sample preparation, the analysis of the test is difficult due to the uncertainties in assessing the localization of the onset of debonding. Moreover, the slicing process leads to a significant relaxation of the residual stresses that could modify the mechanical response of the fiber/matrix interface.

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Figure 3.8: (a) Push-out test load-displacement curve. (b) Scheme of debonding process at different stages. Adapted from Rodríguez et al. (2012b)

The push-in test

Similar to the push-out test, the push-in is performed on the composite cross section directly extracted from the real laminate. However, the sample preparation is simple, assuming that the polishing sequence is known, compared to the push-out case.

The push-in test was originally introduced by Marshall 1984, who used an spherical indenter tip. One could think that a tip with either pyramidal or conical shape might be more useful to push the fibers into the matrix, since their sharpness lets the fiber penetrate deeper without any contact with the surrounding matrix. However, using a sharp tips such as the Berkovich, the Vickers or the cubed-corner ones, leads to residual displacements that will mask the onset of fiber debonding. In particular, when indenting with a sharp indenter, the total measured displacement includes a component that corresponds to the penetration of the tip into the fiber surface. For this reason, the general procedure to identify the debonding consisted in step by step test requiring optical inspection after each increment of displacement in order to verify if the debonding took place. Unfortunately, this procedure is extremely time consuming if several fibers are to be indented.

On the other hand, a modern nanoindenter can monitor both the load and the

displacement at the same time and in a continuous way, which means that the onset of debonding can be obtained observing the change of the slope of the load-displacement curve. In addition, the increasing precision of modern nanoindenters allowed the indentation of smaller fibers, such as carbon and glass ones, extending the applicability of the test to FRPs. However, in both cases the load-displacement curve shows a linear relationship when the onset of the debonding occurs and, hence, it makes the identification of the critical load value, that corresponds to the interface strength, unfeasible. To solve the problem, the penetration of the indenter in the fiber surface must be known and removed from the total displacement measurement, which means that fiber elasto-plastic properties need to be known in advance (Kharrat et al., 1997; Molina-Aldareguía et al., 2011).

Both the spherical blunt tips and the sharp pyramidal/conical should be avoided and a flat punch tip usage is recommended instead. If the punch diameter is close to the fiber one, the tip penetration in the fiber surface is negligible and the total load-displacement curve can be used to determine the critical load that triggers the fiber/matrix interface debonding (Figure 3.9).

Figure 3.9: (Fiber-reinforced polymer composite push-in test with flat punch tip. (a) Push-in test load-displacement curve. (b) Displacement before debonding. (c) Displacement after debonding

The shear stress τ , transferred to the interface is related to the normal load P

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applied to the fiber using the shear-lag model (Cox, 1952; Kelly and Tyson, 1965) including the so called "local environment factor", n , (Molina-Aldareguía et al., 2011; Rodríguez et al., 2012a) according to

$$\tau = \frac{nP}{2\pi r_f^2} \quad (3.10)$$

where r_f is the fiber radius. The value of the dimensionless parameter n depends on fiber and matrix elastic properties as well as on their neighborhood and it can be determined from the stiffness of the elastic region in the load-displacement curve, S_0 , as

$$n = \frac{S_0}{\pi r_f E_f} \quad (3.11)$$

where E_f are the fiber Young modulus. Finally, introducing Equation 3.10 into Equation 3.11, the interface shear stress is related to the normal load as

$$\tau = \frac{S_0 P}{2\pi^2 r_f^3 E_f} \quad (3.12)$$

Therefore, when the normal load reaches the critical value required to trigger the debonding, $P = P_c$, the interface strength T_d is obtained from Equation 3.11 as $T_d = \tau_c = \tau(P = P_c)$. However, it was shown by Molina-Aldareguía et al. (2011) that the interface strength obtained with this methodology underestimated the actual properties, particularly in the case of composites with a large fiber volume fraction ($> 50\%$) because it did not take into account accurately the constraint of the surrounding fibers. In particular, neither the influence of residual thermal stresses, originated in the curing process due to the mismatch between the fiber and matrix linear thermal expansion coefficients, nor the anisotropic nature of fibers and the influence of the surrounding fibers in the stress distribution during the push-in test are considered using the simplified shear-lag model (Equation 3.12).

In view of this, Rodríguez et al. (2012a) modified the simplified shear-lag methodology to obtain more accurate values of the interface strength in composites containing large volume fraction of either isotropic or anisotropic fibers. Rodríguez et al. (2012a) assumed that the fiber/matrix interface behavior is independent of the fiber distribution and, thus, interface parameters can be obtained experimentally by selecting the most appropriate homogeneous fiber distribution. Of course, the specific

Figure 3.10: (a) Optical microscope of a cross-section of an AS4/8552 composite ply. (b) Atomic force microscope image of a hexagonal fiber cluster after the AS4 central fiber has been pushed in with a 3 micrometer diameter circular flat punch

load-displacement curve will depend on the exact fiber distribution that control the residual stress state and the fiber environmental constraint, but the exact numerical analysis and the diverse casuistry of fiber locations make this problem difficult to address and simplifications should be carry out. The novel methodology takes advantage of the fact that fibers surrounded by six fibers in a hexagonal packing pattern are very often found in composites containing a large fiber volume fraction (Figure 3.10). In addition, the critical load required to trigger the debonding in the pushed fiber is minimum for this hexagonal packing. This is important to avoid fiber breakage before debonding.

If the push-in tests is always carried out in the central fiber of an hexagonal packing, the constraint nearest-neighbor fibers can be considered as nearly constant. Therefore, the push-in test can be reproduced using a finite element model that can account for the effects of fiber anisotropy, hygrothermal residual stresses and interface friction after debonding. It should also be noticed that the creation of a new free surface in the composite (to carry out the push-in test) leads to a redistribution of the residual stresses generated in the bulk upon cooling from the curing temperature. The clamping residual stresses on the fibers are released at the free surface and fibers tend to protrude from the new free surface of the specimen.

3.3. *Ex-situ* experimental techniques

They are restrained by the development of shear stresses at the fiber/matrix interface that will be of opposite sign to the shear stresses induced by the indenter during the push-in test. These shear residual stress are accounted for naturally in the finite element simulations of the push-in test.

Using the finite element model a parametric study of the influence of the interface shear strength in the critical shear stress required to trigger the debonding is performed. The resulting relationship is used to obtain the interface strength of the tested material introducing the critical shear stress measured in the real push-in tests. This reverse engineering process has been successfully applied by the author to different types of FRPs, including glass/epoxy, carbon/epoxy and carbon/thermoplastic composites (Naya et al. (2017b)). The particular application to the AS4/8552 composite material, including the finite element model set-up and simulation details are explained in the next chapter.

3.3 *Ex-situ* experimental techniques

3.3.1 Single fiber tensile test

Under tension, carbon fibers are assumed to behave as linear elastic transversely isotropic solids until brittle failure takes place. Their behavior under other loading states is unknown, due to the impossibility of performing specific tests. Therefore, the mechanical characterization of carbon fibers is usually limited to the elastic modulus and tensile strength determination. A description of the available testing methods used to obtain the tensile behavior of the carbon fibers can be found along the next paragraphs. Experimental details and results are gathered in the next chapter together with the ones corresponding to the remaining constituents of the composite.

The tensile stiffness of fibers are usually well characterized straining up to failure individual fibers or strands/tows containing thousand of fibers. However, carbon fibers are brittle materials whose mechanical response is controlled by the distribution and size of defects. According to Reynolds and Sharp (1974), the failure is originated due to the concentration of shear stress acting on misoriented graphite planes at initial defects. In view of this, determining the influence of size and orientation of initial flaws seems the way to do in order to reduce the considerable spread in the experimental measurement of the carbon fibers tensile strength.

Unfortunately, this is a tremendous task that cannot be easily accomplished since

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the number of defects per fiber is huge and the number of fibers per composite is even larger. For this reason, the weakest link theory (WLT) is used to model the failure probability of carbon fibers, $F(\sigma, V)$. Therefore, a fiber of volume V subject to uniform tensile stress σ has a failure probability $P(\sigma, V)$ given by the Weibull distribution according to Equation 3.13 (Weibull, 1951), where V_0 is a reference volume used to characterize the fiber and σ_0 is the corresponding reference stress that combined with the reference volume V_0 gives a probability of failure $F(\sigma_0, V_0) = 0.632$. On the other hand, the parameter m is the so called Weibull modulus, a dimensionless number that controls the stress dispersion. Large values of m can be assumed as a sign of good manufacturing quality, since the fiber stress variation decreases when increasing m .

$$F(\sigma, V) = 1 - \exp \left(-\frac{V}{V_0} \left(\frac{\sigma}{\sigma_0} \right)^m \right) \quad (3.13)$$

To obtain the Weibull parameters, individual fibers are carefully extracted from the tow and tested one by one. The gauge length is fixed and the reference volume is calculated. Then, the fiber is stretched up to failure and the fracture stress σ_u is recorded. The test is repeated for N fibers and the failure stresses are arranged in ascending order. The failure probability of each fiber is assigned according to Equation 3.14, where i is the rank position.

$$F_i = \frac{i - 0.5}{N} \quad (3.14)$$

Taking logarithms in Equation 3.13 and setting $\sigma = \sigma_u$, the following linear relationship is obtained,

$$\ln \left(\ln \left(\frac{1}{1 - F} \right) - \ln \left(\frac{V}{V_0} \right) \right) = m(\ln(\sigma_u) - \ln(\sigma_0)) \quad (3.15)$$

At this point, plotting $\ln \left(\ln \left(\frac{1}{1 - F} \right) - \ln \left(\frac{V}{V_0} \right) \right)$ vs. $\ln(\sigma)$ and doing a linear fitting, the Weibull modulus m is obtained as the slope of this linear relationship (Figure 3.11). On the other hand, the abscissa where the linear fitting cuts the horizontal axis is $m \ln(\sigma_0)$, which allows the calculation of σ_0 .

Once the Weibull parameters and, hence, the fiber strength distribution have been determined, another test can be used to determine the mode one fracture toughness K_{IC} . However, the process is by far more complicated since an artificial

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Figure 3.11: Scheme of log-log plot used to obtain the Weibull parameters (σ_0 and m) by means of linear fitting of fiber failure probability experimentally obtained

notch has to be introduced in the fiber in order to use testing methods based on the linear elastic fracture mechanics theory. In particular, if a is the crack depth, σ is far-field tensile stress, K_{IC} is calculated as

$$K_{IC} = Y(a/d_f)\sigma\sqrt{\pi a} \quad (3.16)$$

where $Y(a/d_f)$ is the shape function for the stress intensity factor of the cracked fiber. The factor $Y(a/d_f)$ for a straight crack in a carbon fiber subjected to uniaxial tension along its axis was determined numerically using the finite element method by [Herráez et al. \(2016\)](#).

Using a FIB the initial shape of the notch, in terms of depth and tip radius can be precisely controlled. Therefore, stretching the notched fibers and recording the maximum stress the fracture toughness is calculated using Equation 3.16. Similar to the procedure used to determine the Weibull parameters, the test is performed for several notched fibers that are stretched up to failure.

Micromechanical characterization at the constituent level

4.1 Microstructure analysis

In order to study the microstructure of the AS4/8552 composite, small rectangular pieces ($10 \times 3 \times 0.5 \text{ mm}^3$) were extracted from the laminate plates. A diamond wire was used to cut perpendicular to the fibers in order to obtain samples of the same thickness. At this point, samples were mounted on holders and polished using different water-based suspensions containing a high concentration of polycrystalline diamonds (Struers DP-Paste P) each one of different grain sizes (9, 3 and $1 \mu\text{m}$) and combined with different polishing clothes (Struers MD/DP DUR and MD/DP NAP). The polishing was carried out using a Buehler MetaServ 250 polisher equipped with a semiautomatic power head. The contact force was set to 5 N and the polishing plate rotating speed ranged between 100 and 300 r.p.m. depending on the diamond suspension and polishing cloth. During the polishing process, alcohol-based lubricant (Struers DP-lubricant blue) was incorporated continuously to improve the grinding finishing.

In order to remove the residual moisture as well as the humidity absorbed during the sample preparation, the polished samples were dried in a stove (AFA 200/150 Dycometal furnace) at 60°C . The samples were weighted every day using a precision balance, AG245 Mettler Toledo 0.0001 grams of resolution, in order to check when the drying process was concluded, which means that the difference between two consecutive measurements is below a specific threshold, according to the DIN EN2823 standard. Typically, after 5 days the samples are completely dry.

4.1. Microstructure analysis

At this point, some of the specimens were taken to the lab in order to perform a visual inspection using an optical microscope. Thanks to the careful polishing process, the fibers are clearly distinguished and the microstructure is revealed (Figure 4.1). Looking closer at Figure 4.1, it can be observed how fiber diameters are very similar. The fibers distribution is quite homogeneous, although some cluster and matrix rich regions are present. In addition, this visual inspection reveals the absence of pores.

Using an image processing program, like the open-source Java-based ImageJ, some important information can be extracted from the micrograph. First of all, a threshold filter is applied to split matrix and fibers into two well-differentiated parts (Figure 4.2). At this point, each fibre is isolated individually to measure its diameter. Repeating this action for all the fibers, the diameter distribution is obtained. In addition, the position of the fiber centers are obtained as well. This information is used then to calculate the distribution of distances to the first neighboring fibers and so on. This kind of statistics are used for some algorithms (such as the NNA) in order to generate virtual microstructures that are statistically equivalent to the real one. Both the fiber diameters, the distance to first neighbor and the distance to second neighbor distributions are obtained using a logarithmic-normal fitting. The results are gathered in Table 4.1.

Table 4.1: AS4/8552 statistics used to generate virtual microstructures

Parameter	Distribution	Mean (μm)	Standard deviation
Fiber diameter	Log-normal	7.25	0.41
Distance to 1 st neighbor	Log-normal	7.46	0.35
Distance to 2 nd neighbor	Log-normal	7.84	0.63

Figure 4.1: AS4/8552 cross section. Image taken using an optical microscope at 500x.

Figure 4.2: AS4/8552 cross section. Image taken using an optical microscope at 500x.

4.2 Polymer matrix characterization

Among the different constituents, the polymer matrix plays a key role in order to determine the non-linear response of the composite ply, specially under transverse compression and in-plane shear. For this reason, its elasto-plastic behavior must be accurately predicted. So far, this has been addressed by fitting against experimental results at the ply level, [Pinho et al. \(2006a\)](#); [Melro et al. \(2013a\)](#), or by means of macroscopic tests, [Fiedler et al. \(2001\)](#), rather than measuring the mechanical properties at the micro scale with independent tests, hence, losing desirable information on the properties at particular coupons and regions that could affect the prediction, as those derived from manufacturing conditions.

Regarding available micromechanical testing techniques, the most suitable current option to provide *in-situ* micromechanical measurements of the elasto-plastic properties of polymer matrix in composite materials is instrumented indentation, commonly referred as nanoindentation [Canal et al. \(2012b\)](#); [Rodríguez et al. \(2012b\)](#). However, the constraint effect derived from the high fiber volume fraction affects the measured properties of the matrix, making it stiffer as the indenter tip penetrates into the surface [Hardiman et al. \(2012\)](#). In addition, pile-up and sink-in phenomena produced during the nanoindentation process can make the estimation of the real contact area difficult, leading to erroneous predictions of the elastic modulus E when applying the *Oliver and Pharr* method [Oliver and Pharr \(1992\)](#). When modeling the polymer as Drucker-Prager frictional material, very often the parameters required to model the elasto-plastic behavior cannot be determined at once from a single nanoindentation test [Rodríguez et al. \(2012b\)](#). One way of tackling this problem is macroscopic testing [Rodríguez et al. \(2012a\)](#) of resin coupons. However, this approach requires the manufacturing of *ad-hoc* resin coupons and one cannot ensure that the properties are the same as those of the resin inside the composite material, since the local curing conditions might differ. Additionally, macroscopic compression tests have some limitations, i.e. laborious sample preparation and typically a poor estimation of E .

In this section, a novel methodology based on the combination of nanoindentation with resin micropillar compression tests is presented. The aim of this combined methodology is to determine with high accuracy the polymer elasto-plastic behavior using two techniques that can be applied *in-situ* in the composite material, avoiding any type of macroscopic test dependency. In this case, the elasto-plastic re-

sponse of the 8552 resin is determined through this combined strategy for RT/DRY (room temperature and no relative humidity) environmental condition. Then, instrumented nanoindentation tests are repeated in aged samples at 70°C combined with 85% of relative humidity (HOT/WET). As a result, the polymer behavior is obtained for both RT/DRY and HOT/WET without any kind of macroscopic testing. Finally, a set of macroscopic compression tests are performed using samples made of neat resin in order to provide a crosscheck value for the polymer compression strength obtained from the mechanical testing.

Regarding the micropillar compression, tests can be carried out at different strain rates, temperatures and local aging conditions, allowing the incorporation of environmental and dynamic effects on the resin behavior. To prove this, preliminary pillar compression studies were also carried out at temperatures up to 90°C and different strain rates and the results are compared to those obtained from the instrumented nanoindentation. The ability to measure *in-situ* the local mechanical properties of the resin at different environmental conditions opens the possibility of determining, in an accelerated way, the chemical and thermal aging suffered by the resin in real FRPs subjected to service conditions, providing a complete micromechanical *in-situ* testing methodology which is in line with the multiscale strategy developed by several authors during the last years [LLorca et al. \(2011\)](#); [Lopes et al. \(2015\)](#); [González and LLorca \(2007b\)](#); [Canal et al. \(2012b\)](#).

Since the objective of the work was to study the feasibility of the proposed micromechanical tests to determine the elasto-plastic properties of the resin within a composite material, the nanoindentation and micropillar compression tests were carried out in the resin rich areas of a UD laminate made of Hexcel AS5/8552 prepregs. However, some nanoindentation tests were also carried out on pure 8552 resin bulk samples for validation purposes, as it was done for these resin bulk samples tested by conventional macroscopic compression tests, to directly compare the results to those derived from the *in-situ* micromechanical tests.

4.2.1 Neat resin macroscopic compression

A set of compression tests were performed on prisms of neat 8552 resin using an *Instron 3384 floor model universal testing system*. Regarding the shape of the samples, two rectangular prisms with sizes $5 \times 5 \times 10 \text{ mm}^3$ and $3 \times 3 \times 6 \text{ mm}^3$ were tested, which keeps the aspect ratio (height:length) of 2:1 ensuring uniaxial stress state at the center of the sample while preventing buckling. Samples were cut using a circu-

4.2. Polymer matrix characterization

lar saw and then polished using an special holder in order to improve the face-to-face parallelism. The top and bottom faces of the prisms were coated with a mineral lubricant in order to minimize the friction forces with the *Instron* plates, following a similar procedure as that used by [Fiedler et al. \(2001\)](#). To ensure quasi-static loading conditions, tests were carried out at 0.01 mm/s. In addition, frame compliance was measured, recording the instrument displacement with the Instron plates stuck together, in order to correct the experimental curves according to $u = u^{\text{exp}} - C \cdot P$, where u^{exp} and P are the displacement and the load provided by the Instron and C is the machine compliance.

The Young modulus, E , and the compression strength, σ_{max} were measured by means of macroscopic compression tests, and used for comparison with micromechanical tests. The results in Figure 4.3 show that both specimens with different sizes show similar results in terms of modulus ($E_{\text{m}} = 3.37 \pm 0.26$ GPa) and compression strength ($\sigma_{\text{max}} = 162 \pm 3$ MPa and $\sigma_{\text{yc}} = 97 \pm 7$ MPa) up to $\epsilon = 0.2$. Larger strains imply the loss of uniaxial stress state due to non-linear geometrical effects (sample "barreling"). For this reason, stress-strain curves are only considered up to $\epsilon = 0.2$.

Figure 4.3: Stress-strain curves obtained from macroscopic compression tests for the two different sample geometries shown on the right hand side

4.2.2 Instrumented nanoindentation tests for RT/DRY

Nanoindentation tests were performed on a Hysitron TI 950 TriboIndenter (Hysitron, Minneapolis, USA) equipped with a Berkovich (pyramidal) indenter. A set of 30 indentations were performed at an indentation strain rate $\dot{\epsilon} = 0.07\text{s}^{-1}$. Tests were performed under load control for maximum load values ranging between 500 μN and 5000 μN to assess indentation "size effects".

The bulk 8552 samples were prepared in a similar manner to the composite materials until a suitable smooth surface (roughness $R_a \leq 5\text{nm}$) was obtained for nanoindentation. Indentations were performed at different surface locations to remove any spatial effect in the measurements. More than 200 indentations were performed, at maximum loads, P_{max} , ranging between 500 μN and 12 000 μN (a bigger indentation load was possible in this case as the indentation size was not limited by the space available between fibers). Indentations were also performed at different indentation strain rates $\dot{\epsilon}$ between 0.01 and 1s^{-1} , to assess the strain rate sensitivity of the resin, as this is an important effect that must be considered when comparing the results with the micropillar compression tests. A total of five indentations were carried out for each combination of strain rate and maximum load. Finally, in order to investigate the potential influence of worn-out indentation tips, two additional sets of 25 indentations were repeated using a brand new Berkovich tip. In this case, indentations were conducted at $P_{max} = 12\,000\,\mu\text{N}$ under two different strain rates, $\dot{\epsilon} = 0.05$ and $\dot{\epsilon} = 0.07\text{s}^{-1}$.

Assuming a Poisson's ratio between $0.35 < \nu_m < 0.38$ for the 8552 resin, the nanoindentation experiments yielded values for Young modulus ranging between $4.67 < E_m < 4.79\text{ GPa}$, which is in good agreement with the value provided by the supplier ($E_m = 4.67\text{ GPa}$) resulting from tensile test of macroscopic coupons. From now on $\nu_m = 0.35$ will be assumed.

One of the most important effects that must be considered when indentations are performed on resin pockets found on the composite cross section, is the constraint effect introduced by the surrounding fibers [Hardiman et al. \(2015\)](#). This effect can be alleviated to some degree by indenting on larger resin pockets and reducing as much as possible the indentation size, as shown in Figure 4.4. However, shallow indents tend to overestimate the elastic modulus due to indentation size effects arising from the sample surface, skin effects of the polymer and/or increased uncertainties due to tip defects near the tip apex [VanLandingham et al. \(2001\)](#). Figure 4.5 shows the

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Figure 4.4: (a) SEM image of a cross section of the AS4/8552 composite showing matrix rich regions. (b) Atomic Force Microscope (AFM) image obtained using the topography signal showing a 3D view of a group of nanoindentation footprints and (c) detailed AFM 2D view of a single Berkovich tip footprint on the polymer matrix

changes in Young Modulus and apparent hardness with maximum indentation depth found in this work, indicating the region of interest or indentation window where the aforementioned artifacts were reduced. Minimum values for Young modulus were obtained for a maximum depth ranging between 400 nm and 600 nm, for typical resin pocket as the ones shown in Figure 4.4. The Young modulus and apparent hardness determined for large indentation depths in neat bulk resin samples (where the fiber constraint effect is not present) is indicated by arrows in Figure 4.5, demonstrating a good match with those obtained in the composite cross-section.

As explained before, a set of indentation tests were also performed on bulk resin plates at different indentation strain rates, in order to assess strain rate effects and extract properties under quasi-static conditions. As expected, the Young modulus E remained constant with increasing indentation strain rate, whereas the apparent hardness H_{ap} and the elastic work ratio W_e/W_t increased, as shown in Figure 4.6. This increment in apparent hardness reflects the strain rate sensitivity (SRS) of the epoxy resin, as previously observed by Frontini et al. [Frontini et al. \(2015\)](#). Although the SRS, calculated according to $SRS = dH/d\dot{\epsilon}$, is small (≈ 0.0091), it

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needs to be considered to ensure quasi-static conditions, for comparative purposes with the results of the micropillar compression tests. Figure 4.6 also plots the results obtained in the AS4/8552 composite at an indentation strain rate $\dot{\epsilon} = 0.07 \text{ s}^{-1}$, showing identical results to those obtained in the bulk resin plates.

Once the "indentation size" and strain rate effects have been considered, the indentation results ($E = 4.75 \text{ GPa}$, $H_{\text{ap}} = 0.301 \text{ GPa}$ and $W_e/W_t = 0.5441$) can potentially be used to estimate upper and lower bounds for the compressive strength of the polymer resin, as explained in section 3. Since both σ_{yc} and β , cannot be determined simultaneously from nanoindentation, in the case of epoxy resins with aeronautical grade, it is reasonable to assume that β will lay between 22° and 36° Rodríguez et al. (2012b). For this range of β , the master curves (Figure 3.2b) result in a $\sigma_{\text{yc}}(\beta)$ ranging between $\sigma_{\text{yc}}(\beta = 36^\circ) = 152 \text{ MPa}$ and $\sigma_{\text{yc}}(\beta = 22^\circ) = 198 \text{ MPa}$. This range is in agreement with the compressive yield strength of 162 MPa , determined by macroscopic testing. In the following, the nanoindentation results will be combined with micropillar compression tests, in order to give a better estimate of the elasto-plastic properties of the resin, based entirely on micromechanical techniques than can be used *in-situ* on the composite material.

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Figure 4.5: Young modulus and apparent hardness as a function of maximum indentation depth obtained from indentations performed on the composite cross section. As reference, the values obtained on the neat 8552 resin are marked by arrows.

Figure 4.6: Apparent hardness and work ratio as a function of indentation strain rate obtained on the neat resin samples. For comparison, the values obtained on the composite cross section, for indentations performed at $\dot{\epsilon} = 0.07 \text{ s}^{-1}$, are also marked by arrows.

4.2.3 Micropillar compression tests

A FEI Helios NanoLab DualBeam 600i equipped with a Focused Ion Beam (FIB) was used to manufacture micro-pillars on the resin pockets of the composite cross-section (Figure 4.7a). Pillars of 5 μm in diameter with aspect ratios between 2:1 and 3:1 were fabricated using annular milling which produced pillars with a slight taper (4°) (Figure 4.7b). Ion currents were chosen in order to produce suitable pillar geometries while keeping reasonable milling times that minimize Ga^+ ion damage, down to a final current $I = 0.43 \text{ nA}$ - that produced pillars with diameters around 5 μm . For this final step, a gas assisted milling deposition process using $(\text{MnSO}_4) \cdot 7\text{H}_2\text{O}$ was employed in order to improve the surface finish and geometry of the pillars.

Samples were then mounted in a special holder designed for easy transfer between the FIB and the nanoindenter instrument. Compression experiments were conducted using the Hysitron TI 950 TriboIndenter equipped with a 10 μm diameter flat punch tip. Due to the highly non-linear behavior of the polymer matrix, the experiments were conducted under displacement control at a constant displacement rate of $\dot{h} = 40 \text{ nm s}^{-1}$. Considering that the height of the pillars was between 10 μm and 15 μm all the test were performed under quasi-static conditions. Samples were kept in the nanoindenter during 1 hour prior to testing in order to minimize thermal drift between the sample and the tip. A total of 10 pillars were compressed on the resin pockets.

In order to investigate the influence of pillar size and geometrical imperfections in the prediction of mechanical properties, pillars of different diameters, D_0 were also prepared on bulk polymer samples. Each pillar was milled individually adjusting the milled areas as well as the Ga^+ current to keep the aspect ratio $H/D_0 \approx 3$ that is large enough to ensure uniaxial stress state at the center of the pillar while minimizing buckling during pillar compression.

Figure 4.8a plots true stress-strain curves obtained from the 5 μm micropillar compression tests, showing a fair reproducibility, and the typical shape encountered for the macroscopic tests. However, an accurate determination of the polymer modulus from the initial elastic loading requires the application of several corrections in order to take into account the taper of the pillars (β), the pillar base rounding (ρ), and the compliance of the base material, as schematically illustrated in Figure 4.8b).

Unlike it was pointed out in 3, the correction proposed by Sneddon (Fei et al.,

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Figure 4.7: (a) SEM Image of an AS4/8852 ply transverse cross section showing a micropillar milled on a resin pocket . (b) Close up SEM image of a milled pillar.

Figure 4.8: (a) Stress-strain curves obtained from 8552 micropillar compression tests performed on the composite cross-section and (b) schematic of the pillar compression test showing the geometrical parameters involved in the calculation of the material mechanical response

2012) cannot be applied here since the substrate and the pillar are made from the same material. Therefore, in order to account for this together with the geometrical effects, the elastic modulus was determined using Equation 4.1 proposed by Yang et al. (2009), which has already been successfully applied to epoxy micropillar compression tests by Wang et al. (2012).

$$E_{pillar} = \Psi \left(\frac{H}{D_0}, \theta, \frac{\rho}{D_0} \right) \left(1 + \frac{\pi(1-\nu)D_0}{8H} \right) \left(\frac{4H}{\pi D_0(D_0 + 2H \tan \theta)} \right) \frac{dP}{dh} \quad (4.1)$$

Here, $\Psi(H/D_0, \theta, \rho/D_0)$ is a dimensionless function derived from FE simulations (Yang et al., 2009), H and D_0 correspond to the pillar height and top diameter respectively, θ is the taper angle and ρ is the radius of curvature at the base of the pillar (Figure 4.8b). Pillar dimensions were obtained from high resolution SEM images prior to testing. Finally, ν is the Poisson ratio and dP/dh is the slope of the load-displacement curve in the elastic regime.

Additionally, direct comparison of the micropillar results (Figure 4.8a) with the macroscopic tests (Figure 4.3) revealed that the flow stress values were typically larger in the case of the micropillar tests. This reveals a pillar size effect, that has been encountered before in epoxy micropillars Wang et al. (2012) and that has been attributed to surface effects due to the interaction of the resin with the Ga^+ ions during micropillar fabrication. Indeed, inspection of the deformed pillars in the SEM revealed the existence of a stiff skin on the surface of the micropillars (Figure 4.9a), that forms as a result of Ga^+ implantation during pillar fabrication and that delaminates and buckles during the micropillar compression test. This FIB-induced skin (Figure 4.8b) has a strong influence in the polymer micropillar compression, because its stiffness is larger than that of the polymer Wang et al. (2012). Assuming a constant thickness and elastic modulus for the stiff skin, the pillar can be considered as a composite material, where the polymer core is wrapped around by the FIB-induced skin. Finally, under isostrain conditions, the modulus of the polymer can be determined as follows:

$$E = E_{\text{polymer}} = \frac{E_{\text{pillar}} - V_s E_s}{1 - V_s} \quad (4.2)$$

$$V_s = 4 \left(\frac{t_s}{D_0} - \left(\frac{t_s}{D_0} \right)^2 \right) \quad (4.3)$$

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Where V_s is the skin volume fraction t_s is the thickness of the FIB-induced skin. Finally:

$$E = \frac{E_{\text{pillar}} - 4E_s \left(\frac{t_s}{D_0} - \left(\frac{t_s}{D_0} \right)^2 \right)}{\left(1 - 2\frac{t_s}{D_0} \right)^2} \quad (4.4)$$

According to Wang et al. (2012) the modulus of the FIB-induced skin can be estimated analyzing the buckling of a uniform stiff elastic skin attached to a compliant elastic matrix. Since the FIB current and the resin system were similar to the ones used by Wang et al. (2012), a value of $E_s \approx 30$ GPa was assumed for the stiff skin. High resolution SEM images of the tested pillars revealed that in a few of the tests, the skin came off and a direct measurement of the skin thickness was possible, as seen in Figure 4.9a. Moreover, the thickness of the FIB-induced skin could also be estimated by measuring the wavelength of the wrinkles produced on the pillar surface during pillar compression (Figure 4.9b) as it was done by Wang et al. (2012). The average skin thickness was found to be $t_s = 18.4$ nm.

This FIB-induced skin not only stiffens the pillar but also increases its yield limit, σ_{yield} and maximum stress, σ_{max} . Assuming an elastic response of the skin when yielding starts, the load supported by the skin can be calculated as

$$P_s|_{\text{yield}} = \sigma_s|_{\text{yield}} A_s \quad (4.5)$$

, where

$$A_s = \pi(t_s D_0 - t_s^2) \quad (4.6)$$

is the cross section of the skin.

Assuming that the yield point corresponds to the moment when the relationship between the load and the displacement becomes non linear, the load supported by the polymer core at the yield point, $P_{\text{polymer}}|_{\text{yield}}$, can be calculated removing the skin contribution from the yield load measured in the pillar compression tests, $P_{\text{pillar}}|_{\text{yield}}$, according to

$$P_{\text{polymer}}|_{\text{yield}} = P_{\text{pillar}}|_{\text{yield}} - P_s|_{\text{yield}} = P_y - P_s = P_y - \frac{E_s A_s h_y}{H} \quad (4.7)$$

Figure 4.9: (a) SEM image of a detached FIB-induced skin on a polymer resin micropillar after compression. (b) SEM image of FIB-induced skin characteristic wrinkles produced during pillar compression.

, where h_y is the micropillar compression displacement at the yield point. Finally, the yield stress of the polymer core can be approximated as

$$\sigma_{\text{yield}} = \frac{4(P_y - P_s)}{\pi(D_0 - 2t_s)^2} \quad (4.8)$$

In this work, an attempt to correct the effect of the FIB-induced surface skin was made by compressing pillars of different diameter, from 2 to 9 μm, and by assuming a constant thickness of the skin, as all pillars were fabricated using similar milling conditions, using the analysis described above by Wang et al. (2012). Figure 4.10 plots the elastic modulus determined using Equation 4.4 as a function of (a) pillar top diameter and (b) skin volume fraction. Red squares correspond to micropillars tested on composites, while black diamonds represent those tested on bulk resin plates, showing identical results. The results indicate that Equation 4.4

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works reasonably well, but it is not able to account completely for skin effects, as the computed elastic modulus increases slightly with reduction in pillar diameter. In addition, for large pillars the average value of the Young modulus drops to 3.8 GPa. However, all values lay in between the elastic modulus determined by macroscopic compression ($E = 3.3$ GPa) and nanoindentation ($E = 4.75$ GPa) and the average value is 4.11 ± 0.4 GPa which is still reasonably close to the value provided by the manufacturer ($E = 4.67$ GPa). Considering the large experimental uncertainty involved on measuring the elastic modulus in compression due to potential misalignment between the compression plates and the specimen surface, the values obtained using the different testing methods agree reasonably well.

It is worth to mention that the determination of the Young modulus using Equation 4.1 is strongly affected by the measurement of the pillar height. In view of this, a sharp fall of E is observed in Figure b for large pillars ($D_0 > 6 \mu\text{m}$), due to the uncertainty measuring the pillar height derived from the difficulties associated to the manufacturing process. Therefore calculating the average value of the Young modulus for small pillars ($D_0 > 6 \mu\text{m}$) provides a value of $E = 4.33 \pm 0.3$ GPa. This is a much better prediction which is in very good agreement with both the value provided by the supplier and the one determined by means of nanoindentation tests. From now on, this value is used in the test comparison.

Figure 4.11 plots the yield stress, as determined from Equation 4.8 as a function of (a) pillar diameter and (b) skin volume fraction. Again, the red squares correspond to micropillars tested on composites, while black diamonds represent those tested on bulk resin plates, showing identical results. The horizontal line represents the yield stress determined from macroscopic compression tests (black-dotted line). Again Equation 4.8 makes a fair job on accounting for the effect of the skin, but it does not completely account for the observed size effects. This is likely due to the fact that the FIB-induced layer behaves like a thin-walled pipe constraining the radial expansion of the pillar and artificially increasing the value of σ_{yield} , due to the influence of hydrostatic stresses. In this case, extrapolation to zero skin volume fraction, in a plot of σ_{yield} v.s. V_s , as shown in Figure 4.11, gives a yield stress value σ_{yield} of 114 MPa, once again in fair agreement with the macroscopic compression tests.

Finally, Figure 4.12 plots the maximum stress σ_{max} as a function of (a) the pillar diameter and (b) the skin volume fraction. The results are similar to those observed for σ_{yield} , but the size effects are considerably bigger, likely due to the larger constrain

Figure 4.10: Determination of Young's modulus (E) against micropillar top diameter (D_0) and skin volume fraction (V_s). Comparison between micropillars carved on composite cross section (red squares) and neat resin plates (black diamonds).

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Figure 4.11: Determination of yield stress (σ_{yc}) against micropillar top diameter (D_0) and skin volume fraction (V_s). The values of σ_{yc} from macroscopic compression tests (black-dotted line) are shown for comparison. Comparison between micropillars carved on composite cross section (red squares) and neat resin plates (black diamonds).

effect imposed by the FIB skin layer for larger plastic strains. Again, it is possible to extrapolate the value of $\sigma_{\max}$ to zero skin volume fraction, V_s , as shown in Figure 4.12b, yielding a value of $\sigma_{\max} \approx 165$ MPa, which is in very good agreement with macroscopic compression tests. It is worth to mention that this is the result that will be used to be compared against the σ_{yc} value obtained from the instrumented nanoindentation tests.

In summary, the micropillar results are strongly affected by pillar size effects arising from a stiff skin, with a thickness of several tens of nanometers, that develops on the surface of the pillars due to interaction of the polymer with the Ga^+ ions. By milling micropillars with varying diameter, and extrapolating the results to zero skin volume fraction, the elastic modulus, yield stress and maximum stress determined by micropillar compression agree reasonably well with the results of macroscopic compression tests, as shown in Table 4.2.

4.2.4 Novel combined methodology

Both micropillars and nanoindentation tests offered similar results for E . However, as the polymer internal friction angle, β , is unknown, $\sigma_{\max}$ cannot be unequivocally determined using nanoindentation results, as explained in section 2. Fortunately, the $\sigma_{\max}$ obtained from micropillar compression can be used here, in combination with the apparent hardness H_{ap} and the work ratio W_e/W_t obtained from nanoindentation tests, to determine β , using the master curves of Figure 3.2, developed by Rodríguez et al. (2012b). The resulting polymer internal friction angle is $\beta \approx 31^\circ$. Predicted values of E and $\sigma_{\max}$ are gathered in Table 4.2. The modulus value of ≈ 4.75 GPa obtained by nanoindentation and micropillar compression is larger than the 3.3 GPa value obtained from macroscopic tests. This low value of E value might be associated with imperfections/misalignments leading to large errors in the estimation of the modulus. This low value is far from the E value (4.67 GPa) reported by the manufacturer obtained from macroscopic tension tests. However, a good estimation of $\sigma_{\max}$ was obtained from the micropillar compression tests, in good agreement with the macroscopic compression tests, which in combination with the nanoindentation results, gives a good estimation of the internal frictional angle β .

In this section, it was shown how the elasto-plastic behavior of a polymer matrix can be characterized by means of a combination of two different *in-situ* micromechanical techniques, avoiding any type of macroscopic test dependency. In particular, compression of micropillars carved on the resin rich area of a composite cross

Figure 4.12: Determination of maximum stress ($\sigma_{\max}$) against micropillar top diameter (D_0) and skin volume fraction (V_s). The values of $\sigma_{\max}$ from macroscopic compression tests (black-dotted line) are shown for comparison. Comparison between micropillars carved on composite cross section (red squares) and neat resin plates (black diamonds).

4. Micromechanical characterization at the constituent level

Test method	E (GPa)	σ_{yc} (MPa)	β (°)
Macroscopic	3.3	162	22-36
Nanoindentation	4.75	152-198	22-36
Micropillar	4.33	165	31

Table 4.2: Comparison of 8552 elasto-plastic parameters obtained from the different experimental methods

section proved to be a valid option for multiscale purposes. Compared to nanoindentation tests, micropillars compression tests yield similar, but slightly lower, results for the prediction of E and at the same time overcoming completely the fiber constraint effect. In addition, unlike using the nanoindentation technique, micropillars provide a measurement of E and σ_{yc} directly from the stress-strain curve.

Therefore, combining the work ratio W_e/W_t obtained from nanoindentation curves with the σ_{yc} predicted by means of the micropillar compression tests, β is determined using the master curves developed by [Rodríguez et al. \(2012b\)](#), avoiding the uncertainty associated with Berkovich nanoindentation.

4.2.5 HOT/WET characterization

At this point, some of the composite samples were stored in sealed holders to keep them dry until new testing and the remaining ones were introduced in the environmental chamber (CCK-0/125 Dycometal model) in order to start the aging process at 70°C and 85% of relative humidity.

The small size of the samples compared to the one recommended by the DIN EN2823 standard [Beuth \(1999\)](#) allows an accelerated water uptake, representing one of the main advantages of this micromechanical approach. The specimen weight was again daily measured and the aging was finished when the specimens were fully saturated (typically after 12 days of exposure to the hot/wet environment).

The saturation water uptake for the AS4/8552 samples was $\Delta M_c \approx 1.25\%$ of the dry weight. Assuming that carbon fibers are not affected by the moisture exposure, it can be concluded that all the water was absorbed by the 8552 epoxy resin. Using a simple rule of mixtures, the amount of water absorbed by the polymer is calculated as $\Delta M_m = \Delta M_c / V_f$. In this case, assuming an average fiber volume fraction $V_f = 58\%$, the water was absorbed by the 8552 epoxy resin resulted $\Delta M_m \approx 3\%$.

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Experimental set-up

Once the composite samples were fully saturated they were extracted from the environmental chamber and placed immediately in the Hysitron TI 950 TriboIndenter (Hysitron, Minneapolis, USA). Once again, a Berkovich pyramidal indenter was equipped on the low load module of the Hysitron. However, this time including two new features, a special heating stage and a thermal shield for the indenter.

The heating stage is used to heat the sample at the testing temperature, 70°C in this case, in a controlled manner. To this end, the heating device has an integrated cooling system and is connected to a PID temperature controller. The heater was designed to enable concentrated heating of the sample without affecting other sensitive components, like the load transducer and the motorized stages, but also to reduce thermal drift due to expansion and contraction of the elements in the load frame. The heater consists on a wire embedded in a plate attached to a metallic cylinder in order to produce fast and uniform heating concentrated under the sample. The heater lays on an isolating layer that is in contact with a Copper cooling stage connected to a water cooling line. A ceramic spacer ring reduced heat transfer between the heater and the steel ring that was used to clamp the whole assembly to the main steel plate. Four small ceramic cylinders placed at the corners of the main steel plate are used to connect the assembly to a big ceramic plate to minimize thermal conduction (Figure 4.13).

Regarding the tip shield, since the Hysitron experimental set-up displays a vertical loading configuration, the transducer must be protected from heat (mainly radiation and convection). To this end, the Berkovich tip is surrounded by a copper shield that is actively cooled to control its temperature (Figure 4.13), in a similar manner as it was done for the heating stage. In fact, both the heating stage and the shield are connected to the same closed-loop cooling system, as it is schematically shown in Figure 4.13. It is worth to mention that this system relies on the principle of passive heating of the indenter tip. Heat flows from the heated sample to the colder indenter tip when contact is first established, until some equilibrium is reached. For this reason, in order to minimize the drift, the indenter tip is attached to a shaft made of a special glass-ceramic with very low thermal conductivity and thermal expansion coefficient.

HOT/WET instrumented nanoindentation tests

Since the results of the instrumented nanoindentation performed on the com-

Figure 4.13: Hysitron TI 950 TriboIndenter high temperature experimental setup.

posite cross section can be compared to those obtained from the neat resin testing once this region of interest is determined (Figure 4.5), for the sake of simplicity, indentations are carried out directly on the neat resin coupons instead of on the resin rich areas of the AS4/8552. In particular, 25 indentations are performed under load control at $\dot{\epsilon} = 0.07 \text{ s}^{-1}$, large enough to avoid thermal drift effect and small enough to ensure quasi-static conditions. Regarding the maximum load, $P_{\max} = 12000 \mu\text{N}$, which means that $h_{\max} \approx 1200 \text{ nm}$, large enough to avoid any tip effect.

According to Rodríguez et al. (2012a), the polymer internal friction angle can be assumed as constant for the range of temperature and moisture of the tests. Therefore, the value obtained from the nanoindentation-micropillar combined methodology, $\beta = 31^\circ$ is used here. This value of β , together with the $W_e/W_t = 0.515 \pm 0.003$ ratio and the apparent hardness $H_{\text{ap}} = 256 \pm 5 \text{ MPa}$ obtained from the HOT/WET testing, is used here to determined the value of σ_{yc} using the master curves developed by Rodríguez et al. (2012b) (Figure 3.2b).

Since the average work ratio obtained from the HOT/WET tests, $W_e/W_t > 0.5$,

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the *Oliver and Pharr* method [Oliver and Pharr \(1992\)](#) does a fair job predicting the Young modulus because no further pile-up correction is needed. The main output of the tests is gathered in [Table 4.3](#).

Condition	E (GPa)	β ($^{\circ}$)	σ_{yc} (MPa)
HOT/WET	3.7 ± 0.07	31	139 ± 3

Table 4.3: Measured 8552 elasto-plastic parameters obtained from instrumented nanoindentation under HOT/WET environmental conditions

It is worth to mention that a first set of 10 indentations was carried out at $\dot{\epsilon} = 0.07 \text{ s}^{-1}$ on the resin rich areas of the composite cross section. Although the results were similar to those obtained for the neat resin test, both the average Young modulus and compression strength were larger ($E = 4.28 \pm 0.19$ and $\sigma_{yc} = 155 \pm 14$). However, these tests were performed under load control setting the maximum load to $P_{\max} = 500 \mu\text{N}$ in order to reduce fiber constraint effect. Under these conditions the maximum displacement was around $h_{\max} \approx 200 \text{ nm}$, which means that the measurements can be affected by tip defects ([Figure](#)). Increasing the indentation depth and finding a region of interest as it was done for the RT/DRY case will reduce the difference between the neat resin and the composite test.

High temperature micropillar compression test: preliminary results

Once the micropillar compression tests have been established, in this section we present novel capabilities that can arise from using this technique, to obtain temperature and strain rate dependency of the resin in composite materials. In particular, the effect of temperature and strain rate on E and, specially, on σ_{yc} is investigated. The results are used as cross check for those obtained from instrumented nanoindentation at HOT/WET conditions. To this end, pillars of $D_0 = 5 \mu\text{m}$ are used as the baseline geometry. For the sake of simplicity, the pillars are carved on the bulk polymer resin. However, the same job can be done on the composite cross-section, providing *in-situ* capabilities. This time the Nanotest Platform 3 Instrument (Micromaterials, Wrexham, UK) was used instead of the Hysitron TI 950 TriboIndenter (Hysitron, Minneapolis, USA). The main reason is the lack of flat-punch tip equipped with thermal shield available for the Hysitron. In addition, thanks to the horizontal set-up and the tip active heating system of the Nanotest, the effect of the thermal drift is minimized. Finally, the problem of aligning both the tip and the pillar can be solved easier thanks to the tilting stage ([Figure](#)).

Figure 4.14: Nanotest Platform 3 Instrument high temperature experimental setup.

Tests were carried out at three different temperatures, $T = 25, 60, 90^\circ\text{C}$ and three displacement rates, $dh/dt = 2, 40, 140 \text{ nm s}^{-1}$. At $T = 25^\circ\text{C}$ two more displacement rates $dh/dt = 10, 100 \text{ nm s}^{-1}$ were used. In total, 12 pillars were tested, 6 at $T = 25^\circ\text{C}$, 3 at $T = 60^\circ\text{C}$ and 3 more at $T = 90^\circ\text{C}$. Samples were mounted on a hot stage using a high temperature cement (Omegabond 600). A thermocouple was placed on the sample surface and used to match the temperature between the sample surface and the indenter tip in order to minimize thermal drift. Thanks to the NanoTest independent control of the tip and sample temperatures, the final thermal drift achieved prior testing was $< 0.15 \text{ nm s}^{-1}$.

As shown in Figure 4.15 (a), the computed E was found to be independent of strain rate, as was also found by nanoindentation, which is consistent with the expectation that the modulus should be independent of the strain rate and should only be a function of temperature Herbert et al. (2008). On the other hand, E decreases linearly for increasing values of T , as seen in Figure 4.15 (b).

Figure 4.16 (a) is a plot of σ_{yc} as a function of displacement rate for different temperatures T , where a linear relationship can be clearly observed. Increasing dh/dt leads to larger σ_{yc} , as it was the case for the previous nanoindentation tests,

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due to the strain rate sensitivity of the resin. Larger values of T , resulted in lower values of σ_{yc} for all different strain rates.

In the case of polymers that exhibit shear-controlled deformation behavior, the yield stress obeys thermally activated processes following the well-known Eyring's model, which has been shown to apply in the deformation behavior of epoxy resins (Miller et al., 2012).

Following the work by Frontini et al. (2015), an Eyring plot for the 8552 resin was constructed, representing the σ_{max} divided by the absolute temperature, T , versus the logarithmic strain rate (Figure 4.16 (b)). The plot shows that all data fall into straight lines, following the same stress-activated Eyring flow model expected for epoxy resins. Similar slopes were found for $T = 25^\circ\text{C}$ and $T = 90^\circ\text{C}$. However, a different slope was displayed for σ_{max}/T data at $T = 60^\circ\text{C}$, which can be attributed to a secondary relaxation, very common for this epoxy resin for temperatures ranging between 55°C and 75°C (reference from JP required)

The number of micropillars compressed in these preliminary tests is small compared to the RT/DRY case, and most likely not enough to provide accurate results. However, the same trend compared to the nanoindentation tests is shown. In both cases, the Young modulus and the compression strength decrease with temperature and strain rate. Interpolating the results at $T = 70^\circ\text{C}$, a value of $E \approx 4.0\text{ GPa}$ is obtained for the Young modulus. This result is in good agreement with that obtained from nanoindentation $E = 3.7\text{ GPa}$.

Regarding the compression strength, the values shown in Figure 4.16 correspond to pillars of $D_0 = 5\text{ }\mu\text{m}$. At this point, it is worth to mention that the average value of σ_{yc} obtained for pillars of $D_0 = 5\text{ }\mu\text{m}$ tested at RT/DRY was $\sigma_{yc} = 214\text{ MPa}$, whereas the final compression strength (removing the effect of the FIB-induced skin) is $\sigma_{yc} = 165\text{ MPa}$, which means that $\sigma_{yc}(D_0 = 5\text{ }\mu\text{m}) = 1.3\sigma_{yc}$ due to the presence of the skin.

Applying the same 1.3 correction factor to the pillars tested at high temperature and interpolating for $T = 70^\circ\text{C}$, a value of $\sigma_{yc} \approx 132\text{ MPa}$ is obtained for the compression strength. This value is again in good agreement with the one obtained from nanoindentation at HOT/WET ($\sigma_{yc} = 139\text{ MPa}$). These results clearly show the potential of this technique to evaluate the mechanical response of resins at different temperatures and strain rates, including *in-situ* capabilities.

Figure 4.15: Influence of displacement rate (a) and (b) temperature on the 8552 Young's modulus.

Figure 4.16: 8552 (c) $\sigma_{\max}$ as a function of displacement rate for different temperatures. (d) Eyring plot.

4.3 Fiber/matrix interface characterization

The interface strength is measured here using the fibre push-in technique described in Chapter 3. All the push-in tests were carried out in a Hysitron TI 950 TriboIndenter (Hysitron, Minneapolis, USA) equipped with a flat punch of 5 μm in diameter, which was smaller than the diameter of the AS4 carbon fibers (7 μm). The diameter of the flat punch is large enough to guarantee no plastic residual imprint is produced in the tested fiber and the non-linearity of the load-displacement curve can be attributed to the fiber/matrix interface debonding. Fiber push-in tests were performed on the central fiber of highly-packed fiber clusters with hexagonal symmetry, an arrangement easily found in the three materials. All tests were performed under displacement control at 40 nm s^{-1} , which is slow enough to neglect any strain rate effects on the polymeric matrix but fast enough to avoid thermal drift effects during the test.

4.3.1 Determining the critical load

A typical load-displacement (P-h) curve resulting from a push-in test is depicted in Figure 4.17. The initial non-linear region observed in the small load regime of the P-h curve was attributed to the lack of parallelism between the punch and the fiber surface. Once this region is exceeded, the P-h curve becomes essentially linear with an initial stiffness S_0 that is obtained by the least square fitting in this region. This region extends until the critical normal load P_c , corresponding to the onset of debonding is reached. Once the value of P_c is known, the fiber/matrix interface critical shear stress τ_c can be obtained using the shear-lag theory (Equation).

In previous work, the methodology used by [Rodríguez et al. \(2012a\)](#) was adopted in order to measure the critical normal load P_c ([Naya et al., 2017a](#)). To this end, the intersection of the P-h curve with a straight line that passes from two points of the P-h curve determined from two parallel lines to the initial stiffness S_0 drawn with offset displacements of 2% and 10% is required. More details can be found in [Rodríguez et al. \(2012a\)](#).

This definition (somewhat arbitrary) of the P_c provided very good estimations for the critical load at the onset of debonding. However, this methodology is strongly affected by the region used to estimate the initial stiffness S_0 by means of the least square fitting. In addition, calculating the intersection between the P-h curve and the two lines with offset displacements of 2% and 10% can be problematic if a

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Figure 4.17: Representative load-fiber displacement curve during the push-in test. Adapted from [Rodríguez et al. \(2012a\)](#).

complicated (high degree polynomial, Gaussian, etc.) fitting function is used.

In view of this, a new method for estimating P_c is proposed here. This new method, again arbitrary, is more robust and easier to program in order to automatize the whole fitting process. The process relies again on the initial stiffness S_0 but, instead of the P-h curve it uses its derivative.

Therefore, the first part of the methodology consists on determining a proper value of S_0 . To this end, a set of linear fittings, using the least square method, is performed increasing at each iteration the number of P-h curve points used in the fitting. As a result, an S_0 distribution is obtained. The maximum value of this distribution is adopted as the average $\bar{S}_0$ initial stiffness.

Once $\bar{S}_0$ has been determined it is time to fit the P-h curve and calculate its derivative dP/dh , which represents the instantaneous slope. The onset of non-linear region can be related to the deviation of $\bar{S}_0$ respect to the dP/dh calculating the relative error δx according to Equation 4.9. In this work, the threshold value is set to 1%. This new methodology can be easily automatized, providing a fast and robust prediction of the critical load P_c with a minimum user intervention.

$$\delta x = \frac{dP/dh - \bar{S}_0}{\bar{S}_0} \quad (4.9)$$

This new methodology can be easily automatized, providing a fast and robust prediction of the critical load P_c with a minimum user intervention. However, the value of P_c can change depending on the function and the displacement range used to fit the P-h curve. For this reason, every single curve is fitted several times using polynomials of different degree over a wide range of displacements. Plotting the resulting P_c values in a dispersion graph (Figure) the bad fittings are clearly identified and, hence, discarded. The average between the remaining P_c values is calculated. This resulting $\bar{P}_c$ is considered as the real curve critical load.

Figure 4.18:

4.3.2 Critical shear stress vs. Interface strength

Following the methodology developed by Rodríguez et al. (2012a), a parametric study of the influence of the interface shear strength in the critical shear stress required to trigger the debonding is performed. The geometry of the 3D finite element model of the push-in test is depicted in Figure 4.19. It includes the central fiber of radius r_f surrounded by six nearest neighbor fibers in a hexagonal pattern. The seven fibers are embedded in the matrix material and this central region is surrounded by a ring of a homogeneous medium, which stands for the composite.

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Taking advantage of the radial symmetry, the analysis is carried in a wedge of 60° . The radius of the wedge is $32r_f$ while the model length parallel to the fibers is $150r_f$. It was checked that these dimensions were large enough so that the load-displacement curve during fiber push-in was not influenced by them [Rodríguez et al. \(2012a\)](#). The geometrical model was meshed with solid elements (C3D8 and C3D6) and a cohesive surface interaction was inserted between the central fiber and the surrounding matrix to account for interface decohesion and friction during the push-in test. It should be noted that the central fiber was not in contact with the nearest neighbors. The minimum thickness of the matrix layer in between was $0.02r_f$, and it was selected following the detailed observation of hexagonal fiber clusters in the scanning electron microscope and also attending to convergence problems during the simulations derived from meshing.

Figure 4.19:

The fibers were assumed to behave as thermoelastic, transversely isotropic solids while the polymeric matrix was modeled as an isotropic, elastic solid. Plastic deformation of the matrix was not taken into account because it did not influence the critical stress at the onset of fiber-matrix debonding for this particular hexagonal fiber arrangement. It should be noted, according to [Rodríguez et al. \(2012a\)](#) that matrix plasticity should be included to compute the interface shear strength in

the case of isolated fibers or when the matrix shear flow stress is below 50% of the interface shear strength.

Regarding the fiber/matrix interface, its behavior was characterized by a cohesive crack model, where interface decohesion and crack growth are controlled by the shear interface strength, T_d , and the interface fracture toughness, G (González and LLorca, 2007b; Totry et al., 2010). This cohesive behavior is coupled with a Coulomb model controlled by the friction coefficient μ .

The finite element analyses of the push-in test included two steps. In the first one, the thermal residual stresses were introduced in the model by means of an initial elastic thermal step, in which the temperature was homogeneously reduced from the curing temperature up to the test temperature without external constraints. Due to the mismatch of thermomechanical properties between fiber and matrix, the residual stresses are build up. Afterwards, the push-in test was simulated in the second step by means of the vertical displacement of a rigid, cylindrical flat punch (of the same diameter as the one used in the experiments) on the central fiber. More details can be found in Rodríguez et al. (2012a).

In this virtual push-in test, the interface strength T_d is introduced arbitrarily as an input of the cohesive traction-separation law. Therefore, measuring the critical shear stress τ_c (actually the normal load P_c is measured and τ_c is calculated using the shear-lag theory) for the different values of T_d , the following linear relationship is obtained

$$\tau_c = f(T_d) = aT_d + b\Delta T \quad (4.10)$$

, where ΔT stands for the difference between the curing and the test temperature and A and B are two fitting parameters provided by the finite element simulations. According to Rodríguez et al. (2012a), a and b only depended on the thermoelastic constants of the matrix and of the fibers, which means that neither the friction coefficient nor the fracture toughness have any influence on the measurement of the interface strength. However, friction is included here by setting the friction coefficient to $\mu = 0.4$ in order to improve the converge of the FE model. This value provides load-displacement curves where the post-debonding slope is similar to the one obtained from the real tests. Regarding the toughness, a value of $G = 20 \text{ J/m}^2$, once again following the recommendations of Rodríguez et al. (2012a), was adopted here.

4.3. Fiber/matrix interface characterization

The parametric study is repeated for three different environmental conditions, RT/DRY (Room temperature and dry atmosphere), RT/WET (room temperature and 85% of relative humidity) and HOT/WET (70° C and 85% of relative humidity). The thermoelastic constants used in the virtual push-in test for the different constituents and environmental conditions are gathered in Table 4.4. Carbon fiber are assumed not to be affected by moisture and temperature. The elastic homogenized properties corresponding to the AS4/8552 can be calculated using the Chamis rule of mixtures (Chamis, 1989) and the fiber volume fraction obtained from the microstructure analysis ($V_f = 0.58$).

Material	E_1 (GPa)	E_2 (GPa)	G_{12} (GPa)	G_{23} (GPa)	ν_{12}	α_1 (K ⁻¹)	α_2 (K ⁻¹)
AS4	231	12.97	11.3	4.45	0.3	-0.9×10^{-6}	7.2×10^{-6}
8552 (Dry)	4.75	4.75	1.73	1.73	0.35	52.0×10^{-6}	52.0×10^{-6}
8552 (Wet)	4.75	4.75	1.73	1.73	0.35	1.53×10^{-6}	1.53×10^{-6}
8552 (Hot)	3.7	3.7	1.37	1.37	0.35	1.53×10^{-6}	1.53×10^{-6}

Table 4.4: Thermoelastic properties of the constituents used in the finite element simulations of the push-in test. AS4 properties are obtained from Herráez et al. (2015b). The 8552 mechanical properties are measured in the previous section. The 8552 coefficient of thermal expansion for DRY condition is provided by the supplier, whereas for HOT/WET is calculated as described at the end of Chapter 2

The linear relationships obtained for each environmental condition are plotted in Figure 4.20 together with the analytic shear lag equation. On the other hand, the fitting coefficients a and b are gathered in Table 4.5. As it can be observed ...

Material	Condition	a	b (MPa/K)	ΔT (K)
AS4/8552	RT/DRY	0.6124	0.0354	160
AS4/8552	RT/WET	0.5485	0.0039	160
AS4/8552	HOT/WET	0.4899	0.0068	90

Table 4.5: Equation 4.10 fitting parameters obtained by means of the FE push-in model for RT/DRY, RT/WET and HOT/WET environmental conditions

Figure 4.20: Linear relationships obtained by means of the FE push-in model for RT/DRY, RT/WET and HOT/WET environmental conditions

4.3.3 Experimental results

Regarding the RT/DRY characterization, a total of fifteen push-in tests were carried out. The load-displacement curves resulting from the tests are plotted in Figure 4.21. Once the critical load P_c is obtained, it is transformed to critical shear stress τ_c using Equation 3.12. Finally, the linear relationship derived from the virtual push-in test is used to obtain the interface strength T_d . The results are gathered in Table 4.6.

As it was pointed out in the case of instrumented nanoindentation on the polymer matrix, testing aged samples at $T = 70^\circ \text{C}$ requires a special shielding device in order to isolate the nanoindenter transducer from heat. Unfortunately, instead of the standard transducer used in the polymer characterization, the push-in tests require a high load one that could not be equipped with the thermal barrier at the time of testing. Therefore, due to hardware limitations, using the Hysitron TI 950 TriboIndenter (Hysitron, Minneapolis, USA) with a flat punch of $5 \mu\text{m}$ at $T = 70^\circ \text{C}$ was not possible.

At this point the Nanotest Platform 3 Instrument (Micromaterials, Wrexham, UK) appeared as a promising alternative, since it allows a better control of the

4.3. Fiber/matrix interface characterization

thermal drift. However, since the fiber and the punch diameters are quite similar, the precision provided by the Nanotest was not enough to avoid the contact between the punch and the surrounding fibers during the push-in test.

In view of this, the RT/WET condition was used instead of the standard HOT/WET, previously used for matrix characterization. Therefore, a set of 15 push-in tests were carried out on aged samples at room temperature using the Hysitron nanoindenter equipped with the 5 μm flat punch tip. The load-displacement curves are plotted in Figure , whereas the main test results are gathered in Table 4.7.

It can be observed that the interface strength decreases in the RT/WET condition and this value can be considered an upper value of the strength of the interface at 70° C and 85% of relative humidity. The additional effect of temperature on the interface strength can be estimated using the knock-down factor on matrix compressive strength due to HOT/WET conditions which can be thought to be entirely due to the temperature increase with respect to RT/DRY conditions. Under this assumption, the HOT/WET matrix compression strength would be further reduced $\approx 19\%$ with respect to a RT/WET environment. Therefore, applying this knock-down factor to the critical shear stress τ_c measured during the RT/WET push-in test and using the linear relationship obtained by means of FE for HOT/WET conditions, an approximated value of the interface strength T_d is retrieved. The extrapolated values are gathered in Table 4.8.

Figure 4.21: Load-displacement curves obtained from the push-in tests performed on the AS4/8552 composite cross section at RT/DRY (a) and RT/WET (b) environmental conditions.

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Test	S_0 (N/mm)	P_c (mN)	τ_c (MPa)	T_d (MPa)
1	92.46	77.79	40.95	57.62
2	93.03	80.34	42.94	60.88
3	95.55	80.68	46.07	65.98
4	95.15	76.85	43.78	62.24
5	90.19	75.62	40.46	56.83
6	91.45	77.52	41.44	58.43
7	90.89	79.64	43.17	61.25
8	90.87	78.23	41.85	59.10
9	92.88	80.89	44.44	63.32
10	95.55	80.68	46.07	65.98
11	93.07	73.55	39.29	54.92
12	93.48	78.53	43.08	61.10
13	93.38	74.37	41.13	57.93
14	91.93	75.28	39.57	55.37
15	90.97	76.95	41.00	57.71
Average	92.72 ± 1.72	77.79 ± 2.38	42.35 ± 2.11	59.91 ± 3.45

Table 4.6: Results of push-in tests carried out on the AS4/8552 composite cross section under RT/DRY condition

4. Micromechanical characterization at the constituent level

Test	S_0 (N/mm)	P_c (mN)	τ_c (MPa)	T_d (MPa)
1	116.42	52.39	28.90	53.82
2	129.14	49.17	30.12	56.03
3	119.91	44.05	24.93	46.57
4	118.65	48.95	27.73	51.68
5	123.10	45.99	27.11	50.56
6	108.86	55.93	29.27	54.48
7	118.69	42.44	24.75	46.25
8	115.58	50.14	28.84	53.70
9	131.40	43.82	27.14	50.60
10	106.97	51.87	26.79	49.96
11	112.16	35.20	19.43	36.55
12	107.85	56.11	30.68	57.05
13	122.33	47.62	27.43	51.13
14	125.93	50.29	30.88	57.42
15	112.45	54.06	29.51	54.92
Average	117.96 ± 7.55	48.54 ± 5.60	27.57 ± 2.92	51.38 ± 5.32

Table 4.7: Results of push-in tests carried out on the AS4/8552 composite cross section under RT/WET condition

4.3. Fiber/matrix interface characterization

Test	S_0 ($\mu\text{N}/\text{nm}$)	P_c (μN)	τ_c (MPa)	T_d (MPa)
1	116.42	52.39	23.41	49.04
2	129.14	49.17	24.39	51.04
3	119.91	44.05	20.19	42.46
4	118.65	48.95	22.46	47.10
5	123.10	45.99	21.96	46.08
6	108.86	55.93	23.71	49.64
7	118.69	42.44	20.05	42.17
8	115.58	50.14	23.36	48.93
9	131.40	43.82	21.98	46.11
10	106.97	51.87	21.70	45.54
11	112.16	35.20	15.74	33.37
12	107.85	56.11	24.85	51.97
13	122.33	47.62	22.22	46.60
14	125.93	50.29	25.00	52.30
15	112.45	54.06	23.90	50.04
Average	117.96 ± 7.55	48.54 ± 5.60	22.33 ± 2.37	46.82 ± 6.08

Table 4.8: Extrapolated results of push-in tests carried out on the AS4/8552 composite cross section under HOT/WET conditions

4.4 Carbon fiber characterization

4.4.1 Tensile strength and mode I fracture toughness

The AS4 carbon fiber strength and Young modulus were determined by [Herráez et al. \(2016\)](#) using the methodology described in Chapter 3 to obtain the Weibull distribution parameters σ_0 and m .

In this tests, [Herráez et al. \(2016\)](#) carefully extracted single fibers from the tow trying to avoid twisting or any kind of damage during handling. Then, the fibers were separated individually and both sides of each one are glued to a cardboard using an epoxy resin or a cyanoacrylate adhesive ([González, 2000](#); [Herráez et al., 2016](#)). The distance between these two fixed points represents the gauge length L_0 used to determined the corresponding fiber strength according to the scaled Weibull distribution. In this case this gauge length was set to $L_0=20$ mm. The cardboard containing the fiber was placed on a fiber tensile testing machine (Favimat+, Textechno), whose grips tight the fiber at its extremes (the fixed points). The cardboard was split in two, and hence, all the load was transferred to the fiber during the test. The fiber was then stretched up to failure under displacement control at 1 mm min^{-1} , leading to strain rates of the order of $\dot{\epsilon} \approx 10^{-3} \text{ s}^{-1}$. The machine monitored both the load and displacement during the whole process. A total of 15 fibers were tested. Results obtained by [Herráez et al. \(2016\)](#) are summarized in Table 4.9. More experimental details can be found in [Herráez et al. \(2016\)](#).

Once the Weibull parameters and therefore the fiber strength are determined, the test has to be repeated using notched fibers in order to determine their mode I fracture toughness. The same FEI Helios NanoLab DualBeam 600i (FEI, Oregon, USA) equipped with a Focused Ion Beam (FIB) system used to carve the resin micropillars was used here to introduce the artificial notches in the fibers. In this case Ga^+ ions were accelerated in the FIB system by using 30 kV of potential, with the beam current being adjusted to 80 pA. Straight and sharp notches were introduced perpendicular to the fiber axis with a depth a to diameter D ratio $a/D \approx 0.1$. A total of 15 notched fibers were tested. More experimental details can be found in [Herráez et al. \(2016\)](#).

Assuming that neither the small crack tip radius (≈ 50 nm) nor the possible material modification induced during the FIB milling operations will not excessively affect the fracture behavior of the carbon fibre, the resulting toughness can be considered a good approximation of the real material property under the linear

4.5. Concluding remarks

elastic fracture mechanics (LEFM) premises. The resulting mode I fracture energy G_{IC} is shown in Table , together with the rest of fiber tensile properties obtained by [Herráez et al. \(2016\)](#), namely, the longitudinal Young modulus E_1 , the Weibull strength σ_0 and the Weibull modulus m .

E_1 (GPa)	σ_0 (MPa)	m	G_{IC} (J/m ²)
229 ± 14	3959	6.5	52 ± 2

Table 4.9: AS4 elastic modulus, strength and mode I fracture toughness. Adapted from [Herráez et al. \(2016\)](#)

4.5 Concluding remarks

In this chapter, we have shown how the elasto-plastic behavior of a polymer matrix can be characterized by means of a combination of two different micromechanical techniques, avoiding any type of macroscopic test dependency. In particular, compression of micropillars carved on the resin rich area of a composite cross section proved to be a valid option for multiscale purposes. Compared to nanoindentation tests, micropillars compression tests yield similar results for the prediction of E and at the same time overcoming the fiber constraint effect. In addition, compared to the instrumented nanoindentation technique, micropillars provide a measurement σ_{yc} directly from the stress-strain curve.

Combining the work ratio W_e/W_t obtained from nanoindentation curves with the σ_{yc} predicted by means of the micropillar compression tests, β is determined using the master curves developed by [Rodríguez et al. \(2012b\)](#), avoiding the uncertainty associated with nanoindentation.

Micropillar compression tests were also performed at high temperatures, where the effect of temperature on E and σ_{yc} was established. Since the micropillars are carved directly on the composite cross section, this methodology provides *in-situ* capabilities. Therefore, it can be used as part of a multiscale strategy devoted to the prediction of composite ply properties, leading to easier, quicker and cheaper characterization of particular composite coupons for different environmental and manufacturing conditions compared to the traditional macroscopic tests.

Prediction of matrix dependent ply properties

5.1 RVE model set-up and simulation

In this chapter, computational micromechanics based on the analysis of a statistically representative volume element of the material (RVE) is used to predict the composite transverse and in-plane ply properties. To this end, the RVE is subjected to homogeneous stress states (tension, compression and shear) together with temperature increments to account for the residual thermal stresses coming from the curing process.

The microstructure of the RVE of the unidirectional composite is idealized as a dispersion of parallel and circular fibers randomly dispersed in the polymer matrix. A total number of fibers around 50 is enough to capture adequately the essential features of the microstructure of the material [González and LLorca \(2007a\)](#) while maintaining reasonable computing efforts. Synthetic fiber distributions statistically equivalent to the real ones are generated for the analysis. To this end, several strategies are available in the literature [Trias et al. \(2006\)](#); [Melro et al. \(2008\)](#); [Vaughan and McCarthy \(2010\)](#) being the Random Sequential Adsorption (RSA) algorithm [Segurado and LLorca \(2002\)](#), probably, the most popular due to the easiness to achieve large volume fraction of fiber reinforcement. In this work, the RSA algorithm was compared with the Nearest Neighbor Algorithm (NNA) developed by Vaughan and McCarthy [Vaughan and McCarthy \(2010\)](#) using the relevant microstructure statistical information obtained from micrographs of the unidirectional ply cross section. As shown in Figure 5.1, the results revealed well-distributed fiber microstructures without significant fiber clustering or matrix rich regions. Hence, it can be concluded

5.1. RVE model set-up and simulation

that both algorithms deliver similar microstructures. Both of them are described in detail in chapter 2.

Figure 5.1: (a) AS4/8552 cross-section micrograph.(b) NNA virtual microstructure.(c) RSA virtual microstructure

Considering its reliability and computing speed, the RSA algorithm was preferred in this work. Two-dimensional periodic fiber distributions were generated with the RSA algorithm and extruded along the fiber direction to achieve the final RVE's of the unidirectional composite material. The periodic RVE's were then meshed using isoparametric wedge and brick finite elements for fibers and matrix with full integration at Gauss points (C3D6 and C3D8, respectively, in Abaqus [Dassault Systèmes \(2011\)](#)). Typically, each RVE contains approximately ≈ 40000 elements representing a discretization fine enough to capture the large stress gradi-

ents between neighboring fibers. Node positions on opposite faces of the RVE's are identical in order to apply periodic boundary conditions according to the methodology developed by [Segurado and LLorca \(2002\)](#), already described in chapter 2. Simulations were carried out with Abaqus/Standard within the framework of the finite deformations theory with the initial unstressed state as reference. In order to account for possible microstructure influence, five different RVEs are used in the simulations (Figure 5.2). To this end and in order to turn this strategy into a user-available tool, an ABAQUS/Standard plug-in has been developed. The plug-in has a graphical interface, where the user can choose the fibre distribution to be generated in the ABAQUS/CAE interface. This tool allows an easy way to generate different RVE's for unidirectional fibre reinforced composites subjected to multiple load states, providing an easy pre- and post-processing of the finite element analysis. The RVE geometry, boundary conditions and mesh are automatically generated using the plug-in. The periodic boundary conditions are automatically generated using a Fortran subroutine interacting with the plug-in. The Python-ABAQUS interface allows the easy modification of material constitutive equations, RVE geometry and boundary conditions, and the adaptation to the user's requirements. The plug-in can successfully applied to calculate the transverse and longitudinal properties of the AS4/8552 composite.

The RVE's were initially subjected to a homogeneous temperature drop of $\Delta T = -160^\circ \text{ C}$, corresponding to the variation produced during the curing process from ($T = 180^\circ \text{ C}$) to room temperature ($T = 20^\circ \text{ C}$), hence generating realistic residual stress states in the material before mechanical loading. During this initial thermal step, rigid body motions (translations and rotations) must be blocked, applying the suited displacement constraints in the master nodes.

In a second step, the different homogeneous stress states were introduced by applying the appropriate displacements to the master nodes linked with the periodic boundary conditions [González and LLorca \(2007a\)](#); [Segurado and LLorca \(2002\)](#). The displacement and reactions of these master nodes were used to determine the homogenized stress-strain curves under transverse, shear and combined loads, and to derive the corresponding material stiffness and strength properties.

5.2. Constitutive equations and model input parameters

Figure 5.2: Virtual RVE's generated using the RSA code and used to predict the transverse and in-plane AS4/8552 ply properties.

5.2 Constitutive equations and model input parameters

In this section, the constitutive equations used to model the mechanical response of the different constituents are explained, with special emphasis on the behavior of the polymer and the fiber/matrix interface. In the case of the 8552 resin, a modified Drucker-Prager plasticity model including tension and compression damage is used, whereas the fiber/matrix interface is modeled using a surface-based cohesive interaction coupled with a Coulomb friction model. Along the next paragraphs, the relationship between the material properties, measured in Chapter 4, and the main constitutive parameters is explained. In addition, the role of the remaining material properties, either found in the literature or assumed, is shown as well.

Finally the carbon fibers are, once again, modeled as linear elastic and transversely isotropic solids. It is worth to mention that the fiber mechanical response is defined by using the same seven thermoelastic constants used in the simulation of the virtual push-in test described in chapter 4.

5.2.1 Epoxy matrix

In this work, the epoxy matrix is modeled using the damage-plasticity model, named as concrete damaged plasticity in the ABAQUS/Standard (Dassault Systèmes, 2011) material library. Although, the details of its constitutive equation have been explained in Chapter 2, in this section the input of the different parameters required by the model are outlined.

As it was pointed out in Chapter 2, the damaged-plasticity model requires the definition of uniaxial tension and compression behavior but also the evolution of the yield (or failure) surface and the material degradation. Although a complex calibration from detailed experiments (including triaxial tests) is often required, using the Young modulus E_m , the compression yield limit σ_{myc} and the internal friction angle of the polymer, β , resulting from the in-situ micromechanical strategy explained in Chapter 4, good results can be obtained if the right values are assumed for some of the parameters describing the yield surface evolution and material damage. Along the next paragraphs, each part is explained in detail.

Epoxy uniaxial tension and compression behavior

The model assumes that the uniaxial tensile and compressive response is characterized by damaged plasticity and, hence by the evolution of tensile and compressive equivalent plastic strains, $\bar{\epsilon}_t^{pl}$ and $\bar{\epsilon}_c^{pl}$, respectively. Therefore, the first thing that must be defined is the uniaxial stress-strain response and damage evolution of the epoxy (Figure 5.3).

Under pure tension, the epoxy behaves as an elastic solid until the tension strength value is reached. This point, corresponds to the onset of material degradation, in the form of microcracking. Increasing the displacement lead to continuous nucleation and coalescence of microcracks producing a macroscopic softening of the material that can be observed in the σ_t decay (Figure 5.3a). In this work, the linear decay has been specified (Figure 5.4c).

Under pure compression, the stress-strain is again linear until the compression strength value is reached. However, in this case the epoxy is able to undergo severe plastic deformation, up to a critical value $\bar{\epsilon}_{crit}^{pl}$, before the onset of compression damage. In this work, the value of $\bar{\epsilon}_{crit}^{pl}$ is adjusted to 0.32, based on the work of Fiedler et al. (2001). This point corresponds to the onset of compression damage (crushing). The material collapse is modeled by means of a σ_c decay (Figure 5.3).

5.2. Constitutive equations and model input parameters

It is worth to mention that the value of the compression yield limit σ_{myc} measured in Chapter 4 is used here.

Figure 5.3: Schematic of the stress-strain curves used to model the epoxy uniaxial tension and compression response. Damage modes are highlighted in green.

In both cases, together with the σ_t and σ_c decays, two independent uniaxial damage laws, $d_t = d_t(\bar{\epsilon}_t^{\text{pl}})$ and $d_c = d_c(\bar{\epsilon}_c^{\text{pl}})$ must be specified (Table).

In this work, fracture energy is used to reproduce the polymer postfailure behavior under tension loading. With this approach the concrete’s brittle behavior is characterized by a stress-displacement response rather than a stress-strain response. The implementation of this stress-displacement concept in a finite element model requires the definition of a characteristic length associated with an integration point (see smeared crack in Chapter 2). Abaqus automatically estimates this length, based on the element geometry and formulation (typically, the length of a line across an element for a first-order element). This element length is used together with the polymer fracture energy, which is a material property and an user’s input (Figure 5.4a), to calculate the equivalent stress-strain relationship. Therefore, since a linear decay is assumed, the only input parameters here are the tension strength σ_t^0 and the epoxy fracture energy G_{IC} .

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Using this stress-strain relationship and the linear damage evolution law $d_t = d_t(\bar{\epsilon}_t^{\text{pl}})$ shown in Figure 5.4b, Abaqus automatically converts the cracking strain to “plastic” strain using the equations shown in Figure 5.4d.

Figure 5.4: Schematic of (a) stress-displacement curve used to defined the postfailure uniaxial tension response of the epoxy matrix (b)linear damage evolution law (c) resulting stress-strain relationship and (d) set of equations used to convert the cracking strain into the ”plastic” one.

Under compression loading, the stress-strain behavior is provided as a tabular function of inelastic (or crushing) strain $\bar{\epsilon}_c^{\text{in}}$ instead of plastic strain $\bar{\epsilon}_c^{\text{pl}}$. This compressive inelastic strain is defined as the total strain minus the elastic strain corresponding to the undamaged material $\bar{\epsilon}_c^{\text{in}} = \epsilon_c - \epsilon_{0c}^{\text{el}}$, where $\epsilon_{0c}^{\text{el}} = \sigma_c/E_0$. Similar to tensile loading case, Abaqus transforms the inelastic strain to plastic strain according to

$$\bar{\epsilon}_c^{\text{pl}} = \bar{\epsilon}_c^{\text{in}} - \frac{d_c}{(1-d_c)} \frac{\sigma_c}{E_0} \quad (5.1)$$

5.2. Constitutive equations and model input parameters

In this case, a bilinear compression stress decay is used to model the crushing process together with a linear damage evolution law $d_c = d_c(\bar{\epsilon}_c^{\text{pl}})$ (Table 5.1).

Table 5.1: Uniaxial stress-strain curves and damage evolution laws used in the damage-plasticity model.

Uniaxial tension			Uniaxial compression		
σ_t	$\bar{\epsilon}_t^{\text{in}}$	d_t	σ_c	$\bar{\epsilon}_c^{\text{in}}$	d_c
σ_{ymt}	0.0	0.0	σ_{ymc}	0.0	0.0
$0.01\sigma_{\text{ymt}}$	$2G_{\text{IC}}/\sigma_{\text{ymt}}$	$\max\{d_t\}$	σ_{ymc}	$\bar{\epsilon}_{\text{crit}}^{\text{pl}}$	0.0
			$0.1\sigma_{\text{ymc}}$	$2\bar{\epsilon}_{\text{crit}}^{\text{pl}}$	$\max\{d_c\}$
			$0.01\sigma_{\text{ymc}}$	$10\bar{\epsilon}_{\text{crit}}^{\text{pl}}$	$\max\{d_c\}$

The maximum damage under both compression and tension is set to $\max\{d_t\} = \max\{d_c\} = 0.9$ to avoid severe convergence problems. It is worth to mention that, if neither d_t nor d_c are defined, the model behaves as a plasticity model and, hence, $\bar{\epsilon}_t^{\text{pl}} = \bar{\epsilon}_t^{\text{ck}}$ and $\bar{\epsilon}_c^{\text{pl}} = \bar{\epsilon}_c^{\text{in}}$.

Plastic flow and yield surface evolution

So far, the behavior under uniaxial tension and compression has been explained. However, in order to predict the plastic flow and the yield function evolution the damaged-plasticity model requires five additional inputs, namely the dilatancy angle ψ , the eccentricity parameter ϵ , the initial biaxial and uniaxial compression strength ratio σ_{b0}/σ_{c0} , the tensile and compressive meridian yield condition ratio K_c and the viscosity parameter μ_c .

Regarding the yield function (see Equation 2.33) together with the stress invariants J_2 and I_1 , two constants, α and γ , and two functions $B(\bar{\epsilon}^{\text{pl}})$ and $\epsilon_{yc} = \epsilon_{yc}(\bar{\epsilon}_c^{\text{in}})$ are required. The invariants are calculated using the stress tensor $\boldsymbol{\sigma} = (1-d)\bar{\boldsymbol{\sigma}}$ and the remaining parameters are calculated by Abaqus using the first four additional inputs above mentioned.

In the case of the constants, α and γ , the former is calculated using the the initial biaxial and uniaxial compression strength ratio σ_{b0}/σ_{c0} and Equation 2.35, whereas the latter is directly obtained from the tensile and compressive meridian yield condition ratio K_c using Equation 2.36. On the other hand, $B(\bar{\epsilon}^{\text{pl}})$ is obtained by means of Equation 2.34 and the uniaxial stress-strain curves previously defined. Finally, the dilatancy angle ψ and the eccentricity parameter ϵ are used to calculate

5. Prediction of matrix dependent ply properties

the plastic flow G , defined in Equation 2.38, and the constant μ_c is introduced to stabilize the model during damage and improve the convergence (Duvaut and Lions, 1976).

It is worth to mention that α can be expressed in terms of the polymer internal friction angle β according to $\tan(\beta) = 3\alpha$. Therefore, the value of β obtained in Chapter 4 is used here to calculate α and hence the input parameter σ_{b0}/σ_{c0} .

In the absence of triaxial test data, maximum plastic dissipation (associated flow in the meridian plane) is used in this work. Therefore the dilation angle ψ is set equal to the internal friction angle ($\psi = \beta$). Due to similar reason, the eccentricity parameter, that defines the rate at which the flow potential tends to a straight line (linear Drucker-Prager) is left as default $\epsilon = 0.1$. Finally, in order to have associated flow in the deviatoric plane, $K_c = 1$. The five input parameters related to the plastic flow and yield surface evolution are gathered in Table 5.2.

Table 5.2: input parameters related to the plastic flow and yield surface evolution used in the damage-plasticity model.

ψ	ϵ	σ_{b0}/σ_{c0}	K_c	μ_c
β	0.1	$\frac{\tan \beta - 3}{2 \tan \beta - 3}$	1.0	0.00001

Just to summarize, the epoxy elasto-plastic behavior is represented in this work using a damage-plasticity model based on the classic Drucker-Prager plasticity criterion. Regarding the parameters measured in Chapter 4, the elastic modulus E_m is used here to represent the elastic response, the compression strength σ_{myc} is used to determined the onset of the plastic regime under compression loading and the polymer internal friction angle β is introduced in the yield surface evolution though the biaxial/uniaxial compression ratio $\sigma_{yb0}/\sigma_{yc0}$. The remaining parameters are either obtained from the literature or left as default values. Nevertheless, all of them are gathered in section 5.2.3 together with the rest of the properties used in the numerical models and corresponding to the remaining constituents.

5.2.2 Fiber/matrix interface

The debonding between fibers and matrix is modeled by means of a classical cohesive zone method coupled with frictional behavior. The constitutive equation that couples both phenomena relates the displacement jump across the interface (Δ) to the traction vector acting on it (τ) by means of a single normalized scalar damage

5.2. Constitutive equations and model input parameters

variable (d) to simulate delamination and friction under the full range of modes (normal - τ_N , transverse shear - τ_T , and longitudinal shear - τ_L) and mode mixities:

$$\tau = \begin{Bmatrix} \tau_T \\ \tau_L \\ \tau_N \end{Bmatrix} = (1-d)K \begin{Bmatrix} \Delta_T \\ \Delta_L \\ \Delta_N \end{Bmatrix} - dK \begin{Bmatrix} 0 \\ 0 \\ \langle -\Delta_N \rangle \end{Bmatrix} + \mu dK \begin{Bmatrix} \langle -\Delta_N \rangle \frac{\Delta_T}{\Delta_S} \\ \langle -\Delta_N \rangle \frac{\Delta_L}{\Delta_S} \\ 0 \end{Bmatrix} \quad (5.2)$$

wherein $\Delta_S = \sqrt{\Delta_T^2 + \Delta_L^2}$ and K is a scalar penalty contact stiffness that describes the linear-elastic behavior of the interface prior to damage initiation, and under any compressive normal tractions which are assumed not to cause damage to the interface ($\langle x \rangle = \max\{0, x\}$). Coulomb-type friction is introduced by the coefficient μ . The penalty stiffness is a non-physical numerical parameter that should be large enough to ensure displacement continuity in the absence of interface damage while avoiding convergence difficulties due to ill-conditioned stiffness matrix.

This coupled cohesive-friction approach is implemented in the kinematics of surface contact interactions and is available in ABAQUS/Standard [Dassault Systèmes \(2011\)](#). It is used in this work to include the significant effect of friction occurring during and after fibre/matrix debonding. In the present approach, the shear stresses caused by friction at the interface are ramped progressively and proportional to the degradation of the interface, thus once the interface is fully debonded, the surface interaction is uniquely governed by a pure Coulomb model, $\tau_{T,L} = \mu K \langle -\Delta_N \rangle$. Furthermore, friction also affects the cohesive response as shear stresses are ramped with the interface damage variable, as schematically illustrated in Figure 5.5. This causes an increase of the interface shear resistance that is function of the normal compressive tractions applied on it.

For the pure cohesive response, damage onset is controlled by a quadratic interaction criterion that is a function of the fiber/matrix interface strength values for each of the damage modes (τ_i^0):

$$\left(\frac{\langle \tau_N \rangle}{\tau_N^0} \right)^2 + \left(\frac{\tau_L}{\tau_L^0} \right)^2 + \left(\frac{\tau_T}{\tau_T^0} \right)^2 = 1 \quad (5.3)$$

wherein only positive normal tractions affect the criterion. Once debonding is initiated, the cohesive tractions transferred through the interface decrease linearly to

Figure 5.5: Schematics of the shear response of the damage-friction model for fiber/matrix interfaces.

zero according to the energy-based Benzeggagh-Kenane (BK) damage propagation criterion [Benzeggagh and Kenane \(1996\)](#).

When using the traditional cohesive elements, instead of the surface-based cohesive approach, friction can only be included when the cohesive element is totally damaged and removed from the finite element mesh. The combined effect of friction with cohesive behavior has been addressed by other authors, e.g. [Alfano and Sacco \(2006\)](#); [Guiamatsia and Nguyen \(2014\)](#). These works eventually led to the development of a cohesive element formulation that take both mechanisms into account; a capability similar to the one used in this work with cohesive surface interactions.

Regarding the input parameters, three interface strengths (τ_N^0 , τ_L^0 and τ_T^0), three fracture energies (G_{Ic} , G_{IIc} and G_{IIIc}), the BK η exponent and the friction coefficient μ need to be specified. At this point, the interface strength measurements obtained by means of push-in tests in Chapter 4 are used, setting $\tau_L^0 = T_d$. In the absence of more reliable data, shear strength in the transverse direction is assumed to be equal to the longitudinal one, $\tau_T^0 = \tau_L^0 = T_d$, whereas the normal interface strength is set to $\sigma_N^0 = 2/3\tau_L^0$ according to [Ogihara and Koyanagi \(2010\)](#).

Concerning the interface fracture energies, to the knowledge of the author, there is no reliable experimental method to measure any of them. So far, setting the fracture energy according to parametric studies trying to reproduce macroscopic experimental results seems to be the best option ([Canal et al., 2012b](#)). However, the fracture energy obtained in this way includes several dissipation phenomena (matrix plasticity, fiber bridging, etc) together with the pure interface debonding

5.2. Constitutive equations and model input parameters

process, what, in the case of the normal mode, can overestimate the real fiber/matrix interface fracture energy. It is worth to mention that assuming a value of $G_{IC} = 150 J/m^2$ and a linear softening, the displacement at failure $\delta_f = 2G_{IC}/\tau_N^0$ would be larger than the fiber diameter. In view of this, the mode I interface fracture energy, G_{IC} , is assumed to be in the range of $2 - 5 J/m^2$. Similar values were used by other authors and reported in the literature [Melro et al. \(2013b\)](#); [Vaughan and McCarthy \(2010\)](#).

In the case of shear mode energies, G_{IIc} and G_{IIIc} , the micromechanical hypothesis made for the normal mode is not that clear, because matrix plasticity and friction during interface failure cannot be uncoupled from the debonding process. Therefore, due to the lack of experimental data, the interface fracture energies in the shear modes were set equal to the matrix cracking fracture energy, $G_{IIc} = G_{IIIc} = G_m$. The fracture energy is assumed to be insensitive to the environmental conditions for the sake of simplicity. Finally, a value of $\eta = 1.45$ was adopted for the B-K exponent according to [Lopes et al. \(2009\)](#).

Similar to the epoxy matrix, all the parameters used to model the interface response are gathered in section 5.2.3 together with the rest of the properties used in the numerical models and corresponding to the remaining constituents.

5.2.3 FE model input parameters

In order to capture the influence of different environmental conditions on the ply behavior, the input parameters of the constitutive models correspond to micromechanical characterization on the resin matrix and on the fibre/matrix interface performed at room temperature conditions (RT/DRY), as well as on samples conditioned up to saturation in an environmental chamber at 70°C and 85% relative humidity (HOT/WET). The constituent material properties used in this work are gathered in Table 5.3.

The set of thermoelastic properties for the AS4 carbon fibers is assumed to be independent on the environmental conditions and is constituted by data provided by the material supplier or derived from the literature [Hinton et al. \(2004a\)](#) using the Chamis rule of mixtures [Chamis \(1989\)](#), as it was done in [Herráez et al. \(2015b\)](#). Polymer elastic modulus, internal friction angle and compression strength (E_m , β and σ_{myc}) have been measured by instrumented nanoindentation under RT/DRY and HOT/WET conditions, as it was explained in Chapter 4. The remaining mechanical properties, namely, fracture energy, tensile strength and Poisson ratio (G_m ,

5. Prediction of matrix dependent ply properties

σ_{myt} , ν_{m} , α_{m} and β , respectively) are provided from the material supplier or taken from literature [Herráez et al. \(2015b\)](#). The coefficient of thermal expansion (CTE) under RT/DRY conditions was provided by the supplier, whereas for HOT/WET conditions, it has been modified, to include the effect of water absorption, according to the procedure described at the end of Chapter 2. It is worth to mention that the AS4 and 8552 thermoelastic properties used here are the same used in the FE model of the fiber push-in explained in Chapter 4 (see Table 4.4).

Using the properties gathered in Table 5.3 and the procedure explained in section 5.2.1, the epoxy uniaxial behavior as well as its plastic and damage evolution, required by the damage-plasticity model, are specified for RT/DRY and HOT/WET conditions.

Regarding the fiber/matrix interface, mode II shear strength (τ_L) has been measured by push-in tests under RT/DRY and HOT/WET conditions (Chapter 4), whereas the remaining properties are set according to micromechanical hypothesis explained in the previous section.

Table 5.3: Properties of the AS4/8552 material constituents used in the FE simulations

AS4 carbon fiber properties							
	E_{f1} (GPa)	E_{f2} (GPa)	G_{f12} (GPa)	G_{f23} (GPa)	ν_{f12}	α_{f1} (K ⁻¹)	α_{f2} (K ⁻¹)
	231	12.97	11.28	4.45	0.3	-0.9e-6	7.2e-6
8552 epoxy matrix properties							
Condition	E_{m} (GPa)	ν_{m}	σ_{myt} (MPa)	β	σ_{myc} (MPa)	$\mathcal{G}_{\text{m}}$ (J/m ²)	α_{m} (K ⁻¹)
RT/DRY	5.07±0.3	0.35	121	29	176±17	100	52e-6
HOT/WET	4.28±0.2	0.35	104	29	152±08	100	1.5e-6
AS4/8552 fibre/matrix interface properties							
Condition	σ_N (MPa)	τ_T (MPa)	τ_L (MPa)	$\mathcal{G}_{\text{Ic}}$ (J/m ²)	$\mathcal{G}_{\text{IIc}}$ (J/m ²)	$\mathcal{G}_{\text{IIIc}}$ (J/m ²)	
RT/DRY	42	64	64±2.64	2	100	100	
HOT/WET	30	45	45±2.72	2	100	100	

5.3 Transverse and in-plane shear uniaxial behavior

5.3.1 Transverse tension response

Under pure transverse tension loading, the fracture process is controlled by the fiber/matrix interface debonding, for both RT/DRY and HOT/WET conditions. Cracks start at the fiber poles along the loading direction in those regions where the stress concentrations in the fiber/matrix interface are higher, for instance in a fiber cluster. After failure of the interface, the matrix undergoes severe plastic deformation, accumulating damage until ultimate failure of the matrix ligaments. The final failure of the RVE is produced by the development of a crack perpendicular to the loading axis, as shown in Figure 5.6. The behavior is linear and elastic up to interface failure. At this point, due to the low mode I interface toughness, the resulting matrix ligaments support all the transverse load immediately. Therefore, the transverse tension strength of the composite is strongly controlled by the fiber/matrix interface strength.

Looking at the stress-strain curves (Figure 5.7), it can be noticed that the amount of plastic strain accumulated by the polymer prior to failure is different for RT/DRY and HOT/WET conditions. The origin of this behavior comes from the combination of a relatively high matrix tensile strength with a low interface strength under HOT/WET conditions. Once the interface fails, the resulting ligaments deform accumulating plastic strain and damage and the matrix is able to absorb the load before reaching its tension strength and finally, failing. Since the reduction of matrix tensile strength is lower than the reduction of interface normal strength, when moving from RT/DRY to HOT/WET conditions, the matrix deforms more before failing. Nevertheless, due to the small interface toughness, the matrix enters in the plastic-damage regime very fast resulting in a poor strength increment once the interface fails.

Figure 5.6: Predicted accumulated plastic strain (PEEQ) and failure mode in a AS4/8552 ply for transverse tension

Figure 5.7: Predicted AS4/8552 stress-strain curve under transverse tension for RT/DRY and HOT/WET conditions. Thin line represents a single RVE. Thick lines correspond to the average of all the RVEs.

5.3. Transverse and in-plane shear uniaxial behavior

5.3.2 Transverse compression response

Under pure transverse compression, the final failure of the composite ply takes place by the development of matrix shear bands. However, the fiber/matrix interface plays an important role in the failure initiation process. According to the in-situ nanoin-dentation tests, the nominal shear strength of the AS4/8552 fiber/matrix interface is lower than the shear strength of the 8552 resin matrix, specially under HOT/WET conditions. If these were the only two mechanisms at play, the simulations show that failure under pure transverse compression would initiate by interface decohesion at the fibre poles and then propagate in the form of a plastic shear band oriented at $\theta_{fr} \approx 47^\circ$. The introduction of frictional effects in the interaction between fiber and matrix changes this equilibrium between fiber/matrix debonding and matrix shear banding. Friction leads to the increment of the interface shearing resistance due to the normal compressive stresses at the interface generated by the thermal and transverse compressive loading. For significant values of μ , failure under pure transverse compression appears not to be initiated by interface decohesion but directly by shear banding with orientation $\theta_{fr} \approx 56^\circ$. This failure mechanism transition effect is shown in Figure 5.8. From a simple parametric study, the threshold value of the friction coefficient that modifies the failure mechanism from a single plastic shear band at $\theta_{fr} \approx 47^\circ$ to multiple distributed plastic shear bands at $\theta_{fr} \approx 56^\circ$ is in the range $0.2 < \mu < 0.4$.

Similar to the RT/DRY case, a failure mechanism transition is originated by lowering μ under HOT/WET condition (Figure 5.9). Therefore, it can be concluded that friction plays an important role in the failure process for both environmental conditions. For a relatively high frictional effects ($\mu = 0.4$), the composite can sustain a relatively high level of strain under transverse compression, typically around 4-5%, justified by the high compressive resistance of the matrix. Under RT/DRY conditions, values of shear band orientation similar to the later case were reported in previous research works [González and LLorca \(2006\)](#) and are supported by experimental data [Koerber et al. \(2010\)](#) for similar materials. Nevertheless, this might not be the case in HOT/WET conditions, since the water absorbed by the polymer tends to form micro channels around the fibers reducing the friction coefficient [Huang and Sun \(2007\)](#); [Wang et al. \(2014\)](#). Unfortunately, quantifying this drop of μ is not possible and, hence, the two possibilities should be considered for HOT/WET conditions ($\mu = 0.4$ and $\mu = 0.0$). In view of this, stress-strain curves under HOT/WET condition are calculated for $\mu = 0$ and $\mu = 0.4$ (Figure 5.10).

5. Prediction of matrix dependent ply properties

Looking at the stress-strain curves, the influence of the interface (including friction) is clearly revealed. If the resulting compression strength values are compared to the data reported in the literature ([Marlett et al., 2011](#)), the hypothesis of assuming low values of μ under HOT/WET conditions is perfectly plausible. Therefore, in the absence of more reliable data, $\mu = 0.4$ and $\mu = 0.0$ will be assumed as the baseline configuration for RT/DRY and HOT/WET environmental conditions, respectively.

5.3. Transverse and in-plane shear uniaxial behavior

Figure 5.8: Effect of the friction coefficient, μ , in the transverse compression loading when $\epsilon = 5\%$. Prediction of accumulated plastic strain (PEEQ)

Figure 5.9: Predicted accumulated plastic strain (PEEQ) at $\epsilon = 4.8\%$ for $\mu = 0.4$ and $\mu = 0.0$ under HOT/WET environment

5.3. Transverse and in-plane shear uniaxial behavior

Figure 5.10: Predicted AS4/8552 stress-strain curves under transverse compression for RT/DRY and HOT/WET ($\mu = 0$ and $\mu = 0.4$) conditions. Thin line represents a single RVE. Thick lines correspond to the average of all the RVEs.

5.3.3 In-plane shear response

When pure shear loading is applied to the RVE, different behaviors are found depending on the shearing direction, parallel or perpendicular to the fibers. If shear is applied parallel to the fibers ($\tau_{\parallel}$), the failure mechanism is dominated by interfacial decohesion or by matrix yielding, depending on the interface strength [Totry et al. \(2010\)](#). In the particular case of the AS4/8552 material studied in this work, the interface strength is slightly lower than the matrix shear limit. Thus, fracture is triggered by interface debonding rather than by matrix plasticity, similarly to the pure transverse tension case. As the interface debonds, the matrix holds progressively shear loads and plastic band deformations are formed. On the other hand, if shear is applied in the plane perpendicular to the fibers ($\tau_{\perp}$), the deformation pattern after matrix yielding is different. Once interface debonding initiates, fiber rotation starts resulting in a gradually stiffer response of the composite material.

The in-plane shear response of the composite lamina (τ_{12}) was approximated in this work by averaging the values obtained along the fibers $\tau_{\parallel}$ and perpendicular to the fibers $\tau_{\perp}$, as suggested by [Totry et al. \(2010\)](#). The resulting shear stress-strain curves for RT/DRY and HOT/WET conditions are gathered in [Figure 5.13](#).

Figure 5.11: Predicted AS4/8552 stress-strain curves under transverse compression for RT/DRY and HOT/WET conditions. Thin line represents a single RVE. Thick lines correspond to the average of all the RVEs.

5.3. Transverse and in-plane shear uniaxial behavior

Figure 5.12: Predicted AS4/8552 stress-strain curves under transverse compression for RT/DRY and HOT/WET conditions. Thin line represents a single RVE. Thick lines correspond to the average of all the RVEs.

Figure 5.13: Predicted AS4/8552 stress-strain curves under transverse compression for RT/DRY and HOT/WET conditions. Thin line represents a single RVE. Thick lines correspond to the average of all the RVEs.

5.3.4 Results

The values of the elastic constants (transverse elastic modulus and in-plane shear modulus) as well as the predictions of the transverse tensile strength, transverse compressive strength and in-plane shear strength for a AS4/8552 lamina are gathered in Table 5.4. The reported numerical results are the average of five different random realizations and are in good agreement with experimentally-obtained average ply properties reported in the literature [Marlett et al. \(2011\)](#), specially for RT/DRY conditions.

In addition, the observed failure modes (Figure 5.14) match the patterns experimentally observed ([Canal et al., 2009](#); [Totry et al., 2010](#); [González and LLorca, 2007a](#)). Plotting together the stress-strain curves for transverse tension and compression (Figure 5.15), it can be observed how the quasi-brittle behavior of the epoxy is enhanced by the presence of the carbon fibers.

Finally, it is worth to mention that the influence of the friction coefficient under uniaxial transverse compression was revealed. Different values of μ not only change

5.3. Transverse and in-plane shear uniaxial behavior

the compression strength but also the failure mode (interface or matrix controlled). The importance of friction in the in-plane biaxial loading states is investigated in the next section.

Table 5.4: Numerically-predicted vs. experimentally-obtained elastic constants, transverse and shear strengths for a AS4/8552 ply (ply thickness $t=0.184\text{mm}$)

Property	RT/DRY		HOT/WET	
	Micromechanics	Marlett et al. (2011)	Micromechanics	Marlett et al. (2011)
$E_2(\text{GPa})$	9.2	9.6	8.3	8.4
$G_{12}(\text{GPa})$	4.8	4.8	3.1	2.3
$Y_T(\text{MPa})$	61 ± 3	63.9	36 ± 2	24.1
$Y_C(\text{MPa})$	290 ± 30	268.0	141 ± 7	136.0
$S_L^{0.2\%}(\text{MPa})$	55 ± 1	55.2	34 ± 1	23.2
$S_L^{5\%}(\text{MPa})$	88 ± 3	91.6	55 ± 3	38.0

Figure 5.14: Predicted UD AS4/8552 transverse stress-strain curves for RT/DRY and HOT/WET conditions. Matrix stress-strain curves are plotted for comparison (dotted-line)

5.3. Transverse and in-plane shear uniaxial behavior

Figure 5.15: Predicted failure modes in a AS4/8552 ply. Tensile damage for transverse tension (a), parallel shear (c) and perpendicular shear (d). Compression damage for transverse compression (b)

5.4 In-plane biaxial loading

One of the potential applications of computational micromechanics is the prediction of ply failure envelopes, i.e. the failure loci for the whole range of combined stress states. In this work the focus is put on the prediction of the intersection of the failure envelope with the $\sigma_{22} - \tau_{12}$ stress plane. This is carried by applying different combinations of transverse and in-plane shear loads, as represented in Figure 5.16 for RT/DRY and HOT/WET conditions. The numerical results are compared to the predictions of physically-based Puck ply failure criteria [Puck and Schürmann \(2002\)](#) using as model inputs the AS4/8552 material properties available in literature [Marlett et al. \(2011\)](#) and summarized in Table 5.4.

The effect of friction between fibers and matrix is clearly visible on the shape of the failure envelopes mainly on the transverse compression quadrant. The results reported in this work suggest that fiber/matrix friction controls the transition between interface-dominated failure in pure shear loading to matrix-dominated failure for pure compression loading. Shear hardening under moderate transverse compression has been observed experimentally (e.g. [Koerber et al. \(2010\)](#)) and is predicted by Puck’s criteria [Puck and Schürmann \(2002\)](#). If fibre/matrix friction is omitted, or the friction coefficient is low, no change in the failure mechanism is obtained and no shear hardening is predicted by computational micromechanics as reported in

Figure 5.16: Predicted failure locus in a AS4/8552 ply under combined transverse stress and in-plane shear for RT/DRY and HOT/WET conditions(with and without friction)

5.4. In-plane biaxial loading

other works [Melro et al. \(2013b\)](#); [Camanho et al. \(2014\)](#). Other authors assumed an arbitrarily large fiber/matrix interface strength in order to capture this effect [Totry et al. \(2008b\)](#) leading to unrealistic predictions of the in-plane and transverse tension strengths. The results of a parametric analysis of the effect of the friction coefficient on the shape of the failure envelope for the RT/DRY conditions are shown in Figure 5.4. There is a threshold value of the friction coefficient in the range $0.2 < \mu < 0.4$ that triggers the transition of the fracture mechanism. Increasing μ above this value leads to no significant change in the material response except for the increase in the slope of the shear hardening curve.

The transition between interface-dominated shear failure to matrix-dominated occurs when the interface shear strength, including the friction effects, overcomes the matrix shear strength. As failure of the matrix under compression loading starts to dominate the ply failure process, the fracture angle also starts to change from $\theta_{fr} = 0^\circ$ to approach a typical shear fracture. Figure 5.18 shows how that transition is produced in the $\sigma_{22} - \tau_{12}$ frame with increasing ratios of transverse compression over shear loads.

This whole range of processes of ply fracture under transverse compression ($\sigma_{22} < 0$) can be described by a single criterion from the set of Puck failure criteria for plane stress cases [Puck and Schürmann \(2002\)](#):

$$\left(\frac{\tau_T}{S^T - \eta^T \sigma_n} \right)^2 + \left(\frac{\tau_L}{S^L - \eta^L \sigma_n} \right)^2 = 1 \quad (5.4)$$

wherein the tractions on a possible fracture plane with angle θ are obtained from the components of the stress tensor defined in the material coordinate system (see Figure 5.19) as:

$$\begin{aligned} \sigma_n &= \sigma_{22} \cos^2 \theta \\ \tau_T &= -\sigma_{22} \sin \theta \cos \theta \\ \tau_L &= \tau_{12} \cos \theta \end{aligned} \quad (5.5)$$

S^L and S^T are, respectively, the material shear strengths in fibre and transverse-to-fibre directions while η^L and η^T are the corresponding internal friction coefficients acting on the fracture plane. The prediction of the correct θ_{fr} for each biaxial load ratio requires the criterion to be maximised for the whole range of possible fracture angles. Following this procedure will dictate that the fracture angle evolves from

Figure 5.17: Effect of model parameters on the predicted failure locus in a AS4/8552 ply under combined transverse stress and in-plane shear: a) Effect of the friction coefficient between fiber and matrix; b) Effect of polymer matrix internal friction angle.

5.4. In-plane biaxial loading

Figure 5.18: Predicted fracture angles for different load combinations and RT/DRY conditions ($\mu = 0.4$). The image shows the concentration of accumulated plastic strain (PEEQ) for the different biaxial loading states

$\theta_{fr} = 0^\circ$, for transverse tension and moderate transverse compression, to $\theta_{fr} \approx 53^\circ$ for pure transverse compression. A remarkably similar trend is suggested by the computational micromechanical results presented above and by experimental observations [Koerber et al. \(2010\)](#), with sensible differences being that the increase of θ predicted by Equation 5.4 is continuous and progressive up to $\theta_{fr} \approx 53^\circ$ while in micromechanics the increase is in discrete steps and up to $\theta_{fr} \approx 56^\circ$, a value that matches experimental observations [Koerber et al. \(2010\)](#) more accurately. These differences are likely to be related to the discreteness of the microstructure which is not taken into account in the ply failure model.

The effect of friction on ply failure for transverse compression is also taken into account in Puck's criterion wherein the shear strengths are affected, i.e. increased, by negative normal traction acting on the fracture plane on η^L and η^T proportions (note that Equation 5.4 is only valid for $\sigma_{22} < 0$). For $\theta = 0^\circ$, η^L defines the slope of the shear hardening region in the failure envelope. A linear interpolation of the micromechanical simulations in this range, for a fibre-matrix friction coefficient of $\mu = 0.4$, results in $\eta^L = 0.22$ (see Figure 5.4), a value close to the one experimentally observed by [Koerber et al. \(2010\)](#) ($\eta^L = 0.26$) for a similar CFRP system (IM7/8552). Given these correlations, the parametrically-obtained friction coefficient $\mu = 0.4$ is adopted at this point and used in the following predictions.

For a HOT/WET environment, ply fracture seems to be controlled by interface failure in the whole range of transverse biaxial loads. Hence, no shear hardening should be observed in moderate compression. As the drop of friction coefficient due to water uptake is not perfectly defined, two failure envelopes, corresponding to

Figure 5.19: a) Fracture plane for a ply subjected to transverse compression and in-plane shear; b) Stresses in the fracture plane.

$\mu = 0.01$ and $\mu = 0.4$, are represented in Figure 5.16 in order to establish lower and upper bounds for the failure envelope for HOT/WET conditions.

Given the importance of the hydrostatic pressure in the behavior of polymers and the uncertainty about the model parameters that control its effect, namely the internal friction angle, a parametric study on the variation of this parameter on the behavior of the composite was carried out. The internal friction angle (β) is related to the coefficient of the hydrostatic term of constitutive equation of the epoxy matrix (Equation 2.33) and its variation implies a change in the material biaxial compressive response (Equation 2.35). The influence of the polymer matrix plastic behavior in the global composite microstructure can be assessed by comparing three failure envelopes corresponding to values of $\beta = 22^\circ$, 29° and 36° and a fixed value of the friction coefficient ($\mu = 0.4$), as shown in Figure 5.4. The curve for $\beta = 29^\circ$ corresponds to the material properties gathered in Table 5.3 and represents the baseline configuration. The other values considered herein represent a lower and upper bound for most of epoxy resins Rodríguez et al. (2012b).

As observed in Figure 5.4, the material pure transverse compressive strength (Y_C) and the slope of the shear hardening curve (η^L) increase for $\beta = 22^\circ$. However, this effect is not symmetric, i.e. Y_C and η^L do not significantly decrease for an equivalent increase of β to 36° . In addition, the ply fracture angle for pure transverse compression (θ_{fr}) is kept constant regardless the internal friction angle of the polymer, as shown in Figure 5.20. It appears that decreasing β from the baseline value of 29° results in a pronounced increase of the matrix yield load with a direct effect on Y_C . Indirectly, a stronger matrix allows the increase of the normal stresses on the fiber/matrix interface and, as a consequence of friction, an increase on its shear resistance. In other words, decreasing the friction angle of the polymer increases its compressive strength Rodríguez et al. (2012b) and hence of the friction shear load transferred between the fibers and the polymer. On the other hand, increasing β

5.5. Concluding remarks

above 29° appears not to have a significant impact on matrix compressive yielding and neither on η^L , i.e. the effect of this parameter is nonlinear. Hence, in the absence of more objective information, a value of $\beta \approx 29^\circ$ seems to be appropriate to characterize the 8552 epoxy resin behavior as it results in values of Y_C , η^L and θ_{fr} coherent with experimental data.

The results and correlations made in this section lead to the hypotheses that η^L may be regarded as a ply-homogenized combination of the effects of fibre/matrix interface friction and matrix internal friction, while η^T would better correspond to matrix internal friction only. The confirmation of these hypotheses would require further investigation.

Figure 5.20: Predicted compression damage and fracture angle in a AS4/8552 ply under a pure transverse compressive stress state for different polymer matrix internal friction angles, β

5.5 Concluding remarks

In this chapter, the transverse tensile strength, transverse compressive strength and in-plane shear strength of a unidirectional AS4/8552 lamina under both RT/DRY and HOT/WET environmental conditions have been determined using computational micromechanics. The main parameters of the constitutive equations of the microconstituents, including the fiber/matrix interface and polymer plastic behavior, were obtained experimentally by means of *in-situ* nano-indentation tests. Using only micromechanical properties, the model reproduces the ply stress-strain behavior and fracture mechanisms observed experimentally [Canal et al. \(2009\)](#); [Totry et al. \(2010\)](#); [González and LLorca \(2007a\)](#), both for uniaxial and biaxial stress states. Hence, this chapter suggests that virtual ply characterization tests, based on reliable properties of the microconstituents, can replace the physical experiments, at

least for material screening purposes. These virtual tests provide full control of the composite microstructure and constituent properties, allowing microstructural optimization to be performed in the future [Herráez et al. \(2015a\)](#). Moreover, complex stress states, not possible to be applied experimentally, can be simulated. Finally, this chapter shows that there is a need to improve existing ply failure criteria that rely only on ply properties [Dávila et al. \(2005\)](#); [Puck and Schürmann \(2002\)](#). Without exception, these assume microstructures containing perfect fiber-matrix bonding and do not take into account important micromechanical parameters such as fibre/matrix interface strength and interface friction.

Prediction of fiber dependent ply properties

Fiber Reinforced Polymers (FRP's) are nowadays extensively used in applications where very good mechanical properties are required in combination with weight savings. In most the cases, the specific longitudinal properties are their best selling point, and fast ways to determine those constitute an advantage in the process of design of composites. Despite the large number of analytic models developed to predict the longitudinal ply behavior, the longitudinal deformation mechanisms, including the effect of fiber distribution, polymer plasticity and damage as well as that of the fiber/matrix debonding and friction, are non-trivial to simulate.

The aim of this chapter is to develop a reliable and accurate computational approach, supported by the micromechanical *in situ* measurements of the material constituents carried out in Chapter 4, to predict the fiber dependent mechanical properties of a composite ply (stiffness and strength) under different operating environmental conditions. In the previous chapter, this strategy has been successfully applied to study the transverse and shear behaviors. This bottom-up analysis approach enables, among many possibilities, the in-situ virtual testing of specific regions of the ply, the fast characterization of different combinations of matrices and fibers, the study of micro-defects resulting from deficient curing conditions ([Ashouri Vajari et al., 2014](#)). This leads to the reduction of time and costs involved in new material designs and provides fast screening capabilities that improve material selection in engineering applications.

Although the constitutive equations and the mechanical properties used here are the same used in Chapter 5, the longitudinal ply properties depend on fiber defects and, hence, specific RVEs are required for each failure mode. Therefore, this chapter

6.1. Prediction of longitudinal tension

is divided in two main parts, longitudinal tension and longitudinal compression. Along the next sections, the specific set-up for each specific RVE is explained in detail, trying to highlight the most relevant micromechanical parameters governing each failure mode.

6.1 Prediction of longitudinal tension

6.1.1 Volume dependent defects in carbon fibers

Talking about the mechanical response of a UD FRP composite ply under longitudinal tension means talking about fiber breakage, a process dominated by the nature and size of defects along the fiber volume. As it was pointed out in Chapter 3, the Weibull distribution (Equation 3.13) is the best option to model the failure probability of carbon fibers, $P(\sigma, V)$. In view of this, the statistical nature of the ply failure under longitudinal tension is clear.

Assuming that carbon fibers behave as linear elastic solids until fracture and their strain at failure is much lower than the matrix one, the UD ply longitudinal strength, X_T can be modeled using a simple rule of mixtures:

$$X_T = \sigma_u V_f + \sigma_{ymt}(1 - V_f) \quad (6.1)$$

, where σ_u and σ_{ymt} are the fiber and matrix tensile strength, respectively, and V_f is the fiber volume fraction. The problem is then finished if the fiber tensile strength is known. For example, in the particular case of the AS4/8552, using the Weibull parameter σ_0 measured in Chapter 4 as the average longitudinal fiber strength, $X_T = 2105$ MPa, which is actually very close the value reported by Marlett et al. (2011). However, this results has no physical meaning, it is just a lucky coincidence, because the physical meaning of σ_0 is related to the average size of defects in a volume V_0 . In other words, changing the length of the carbon fibers used in the tests explained in Chapter 4, will change the value of σ_0 and hence that of X_T .

6.1.2 Analytical models: GLS vs LLS

Trying to deal with this problem, an important number of analytical models have been proposed in the past (see Rosen (1964); Zweben and Rosen (1970); Curtin (1991); Hui et al. (1995) among others). Although all of them rely on the Weibull distribution to estimate the failure probability, they follow different approaches. In

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view of this, they can be classified as Global Load Sharing (GLS) and Local Load Sharing (LLS) models.

In a GLS model (Curtin, 1991; Hui et al., 1995), once a fiber is broken, the load is redistributed among all the fibers placed on the fracture plane in an homogeneous way, neglecting local effects arising around the broken fiber. In other words, the fracture of a fiber is independent of where previous fractures have been taken place. Under these conditions, the composite failure process can be understood studying the failure of a single fiber embedded in the matrix under a uniform remote stress σ_f . This problem was approximated by Curtin (1991) and solved Hui et al. (1995). Both models arise similar results if the Weibull parameter m is larger than four, which is our case. In view of this and due to its simplicity, Curtin's models is use here instead of the exact solution developed by Hui et al. (1995) to explain the GLS approach.

Curtin's model states that starting at the point where the fiber is broken, the stress increase linearly from zero up to the uniform remote stress value along a distance called the transference length δ^* (Figure). This distance can be determined applying a simple load balance as

$$\delta^* = \frac{\sigma_f r_f}{2\tau_L} \quad (6.2)$$

, where r_f and τ_L are the fiber radius and the mode II interface strength respectively. According to Curtin (1991), the average stress in the broken fiber, $\bar{\sigma}_f$, can be approximated as

$$\bar{\sigma}_f = \sigma_f(1 - F(\sigma_f, 2\delta^*)) + \frac{\sigma_f}{2}F(\sigma_f, 2\delta^*) \quad (6.3)$$

, where $F(\sigma_f, 2\delta^*)$ represents the accumulated failure probability of fiber of length $2\delta^*$ when the applied stress is σ_f . The value of $F(\sigma_f, 2\delta^*)$ is calculated using the Weibull distribution according to

$$F(\sigma_f, 2\delta^*) = 1 - \exp \left\{ -\frac{2\delta^*}{L_0} \left(\frac{\sigma_f}{\sigma_0} \right)^m \right\} \quad (6.4)$$

Combining 6.3 and 6.4, the average stress hold by the broken fibers is

$$\bar{\sigma}_f = \sigma_f \left[1 - \frac{1}{2} \left(\frac{\sigma_f}{\sigma_c} \right)^{\frac{1}{m+1}} \right] \quad (6.5)$$

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Figure 6.1: Stress distribution in a broken fiber according to a GLS model.

, where σ_c is

$$\sigma_c = \left[\frac{\sigma_0^m \tau_L L_0}{r_f} \right]^{\frac{1}{m+1}} \quad (6.6)$$

Calculating the maximum of $\bar{\sigma}_f$ and adding the matrix contribution, the composite longitudinal strength is predicted as

$$X_T = \sigma_c V_f \left[\frac{2}{m+2} \right]^{\frac{1}{m+1}} \left[\frac{m+1}{m+2} \right] + \sigma_{ymt}(1 - V_f) \quad (6.7)$$

This method, published by Curtin (1991), provides a simple solution to the fiber fragmentation problem corresponding to a GLS model and has been successfully applied to predict the longitudinal fracture of ceramic-ceramic composites. However, Curtin's model represents an upper limit when it is applied to FRPs, where damage is often localized in a narrower band.

In order to improve the longitudinal strength prediction in FRPs, a new type of methods, the LLS, must be used. These models incorporate the influence of a fiber breakage into the failure probability of the adjacent ones. Among them, it is worth to mention the pioneering work of Rosen (1964), who stated that the breakage of a single fiber in a group of N is enough to trigger the failure of the composites. According to Rosen (1964), the composite longitudinal strength can be predicted as

$$X_T = V_f \sigma_0 \left[\frac{L_0}{NL} \right]^{1/m} + \sigma_{ymt}(1 - V_f) \quad (6.8)$$

It is worth to mention that very often, several fiber breakages are required to trigger the catastrophic composite failure. Once a fiber is broken, damage will propagate on the fracture plane through the matrix and fiber matrix/interfaces up to the

neighboring fibers, affecting a particular region of the composite, the local representative volume (LRV). Actually, after the fiber breakage, the load redistribution introduces a stress concentration in the neighboring fibers increasing their failure probability. The composite failure stress can be predicted analyzing the probability of sequential breakage of the second, third, fourth, etc, neighboring fibers [González and LLorca \(2001\)](#). However, this is not an easy task, since the stress concentration factors are affected by the matrix and fiber/matrix interface properties as well as by the fiber distribution and must be calculated often using FE models. In addition, once the number of broken fibers is larger than two, the number of ways they can be arranged increases as well. Increasing the number of broken fibers, the number of combinations blow up leading to the usage of Monte Carlo techniques, which are out of the scope of this work.

In view of this, the idea of this section is to include the statistical nature of carbon fiber breakage into a 3D RVE model similar to the ones used in Chapter 5 and provide a method to obtain a value of X_T regardless of the size L_0 and the corresponding Weibull parameters σ_0 and m . However, unlike the GLS model, the 3D RVE incorporates the influence of a fiber breakage into the failure probability of the adjacent ones, similar to the LLS methods. To this end, as we will see later on, the size of the RVE should be modified depending on the value of σ_0 .

6.1.3 Proposed methodology

In this section, a computational micromechanics methodology, based on a periodic RVE approach, is proposed to obtain the longitudinal tensile strength of a UD composite ply using the constituent properties measured in Chapter 4. In particular, the Weibull parameters are the key properties of the model. Depending on σ_0 , the size of the RVE has to be scaled if a similar X_T value is to be retrieved. If fibers larger than the ones used in the single tensile test explained in Chapter 4 are used, a lower value of σ_0 will be obtained and, hence, using [6.1](#), a lower value of X_T . On the other hand, using shorter fibers in the tensile test, a larger value of σ_0 and hence of X_T will be predicted. In order to solve this problem the usage of an RVE whose size depends on the value of σ_0 is used.

The length of the RVE, W , is set according to the expression for δ^* (Equation [6.2](#)), replacing σ_f by σ_0 , and the fiber strength is scaled, using Weibull distribution (Equation [6.9](#)), according to it. The size of the RVE must be $W \geq 2\delta^*$ to allow complete stress recovery. However, increasing σ_0 too much (using fibers of very small

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L_0), will lead to very large RVEs, making the computation unfeasible. On the other hand, small values of σ_0 , will give small RVEs, where the stress recovery could not be produced. A balance value of σ_0 is then required.

$$\bar{\sigma}_u = \sigma_0 \left[-\frac{L_0}{L} \ln x \right]^{1/m} \quad (6.9)$$

6.1.4 RVE model set-up

A periodic RVE similar to the ones used in the prediction of the matrix dependent ply properties is used here. In other words, the composite ply microstructure is again idealized as a dispersion of parallel, circular elastic fibers embedded in the polymer matrix. In order to capture the longitudinal failure of the microstructure, the fracture of carbon fibers has to be reproduced explicitly. To this end, a set of fracture planes, represented by cohesive surface-based interactions, is introduced in the central part of every fiber. A similar strategy was already used in the past [González and LLorca \(2006\)](#); [Romanowicz \(2012\)](#), but neither cohesive interactions including friction nor real fiber distributions were simulated so far.

Fracture planes are distributed along a distance depending on the Weibull modulus m . The higher the value of m , the shorter the distance where fracture planes are placed. In this case, this distance is set to $50\text{ }\mu\text{m}$, enough to contain the final fracture plane as it will be seen later on. A total of 11 fracture planes are placed on each fiber separated by a distance d equal to $5\text{ }\mu\text{m}$, slightly inferior to the fiber diameter $d_f = 7.1\text{ }\mu\text{m}$ (Figure 6.2). This value of d was set trying to maximize the available computational resources. Increasing the number of fracture planes will blow up the computational cost of the model. Therefore, in this model, fiber portions smaller than d are considered to be free of defects and, hence, can not be broken. Since the number of fibers was around 62 (actually 50 due to periodicity) for each microstructure, this means that a total of 682 fracture planes (550 of them are different after imposing periodicity) are placed inside the RVE. This amount of fibers and fracture planes, large enough to capture the effect of the Weibull distribution in the fiber fracture strength, avoids any significant influence in the elastic response of the microstructure [González and LLorca \(2007a\)](#) maintaining computing time under reasonable values.

Regarding the length of the RVE, it was set to $W = 0.225\text{ }\mu\text{m}$ according to the proposed methodology described above. This length is enough to provide stress

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recovery once a fiber is broken, maintaining a local load sharing (LLS) approach and keeping the computational resources under control. Nevertheless, the number of elements increased up to 1.4 million degrees of freedom requiring the use of important computational resources (the amount of RAM memory was larger than 200 GB). In particular, using an HPC cluster of 48 Intel Xeon E5-2670 cores and GPU acceleration, the calculation required around 72 hours to complete.

Figure 6.2: Periodic 3D RVE used in the longitudinal tension FE model

In view of this, a simplified RVE, representing an hexagonal packing was stretched trying to optimize the set-up. During this small test, an important fact was revealed. For the particular case of the AS4/8552, due to the large difference between fiber and matrix tensile strength, regardless of the fiber strength distribution, the influence of matrix plasticity in the maximum stress borne by the RVE is negligible compared to the influence of the fiber/fiber and fiber/matrix interfaces. Matrix plasticity and damage start being relevant during the post-peak regime. Removing matrix plasticity, the only source of non-linearity is the cohesive contact at fiber/matrix and fiber/fiber interfaces. Unfortunately, even using this simplification, the computational time is still remarkable ($\approx 24\text{hours}$) due to the large amount of contact surfaces present in the RVE. Nevertheless, this time saving was enough to choose the usage of an elastic matrix and focus the analysis in the fiber strength distribution and in the presence of cohesive interfaces.

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6.1.5 Results

The longitudinal stress-strain curves for the AS4/8552 UD ply under both RT/DRY and HOT/WET conditions are gathered in Figure 6.7, revealing a linear behavior almost until the moment of failure. Actually, the curves become non-linear once the first fiber fracture takes place. In addition, although the same fiber properties are used for both RT/DRY and HOT/WET conditions, the composite strength is lower in the second case. Since, plasticity has been suppressed the origin of such discrepancy must come from the lower fiber/matrix interface strength under HOT/WET conditions.

As it can be observed in Table 6.2, the numerical results are in good agreement with experimentally-obtained values found in the literature Marlett et al. (2011) for both environmental conditions. However, there is an overestimation of $\approx 16\%$ in strength that deserves some comments. First, the longitudinal tension test is not easy to perform and there is a large scattering from one coupon to another, corresponding to the stochastic distribution of defects. Actually, the supplier reported a value of $x_T = 2207$ MPa for the same material system. On the other hand, the prediction arising from the micromechanical analysis corresponds to the value of a single RVE and a single Weibull distribution. A more realistic average prediction will be achieved repeating the analysis for different microstructures and fiber strength distributions.

Finally, the number of fibers used in the RVE could be not enough to capture the influence of the Weibull distributions. As an example, in order to predict the X_T value reported in the literature, either by Marlett et al. (2011) or by the supplier, using the model developed by Rosen (1964) (Equation 6.8), the Weibull parameters (σ_0 and m) measured in Chapter 4 and fibers of $0.225\mu\text{m}$, the number of fibers ranges between $\approx 136 - 290$. In other words, 2 – 6 times more fibers are required in the RVE. Unfortunately, this was not possible due to hardware limitations at the time of writing these lines.

It is worth to mention that although the model overestimates the value of X_T , it captures the failure mechanisms, allowing to evaluate the role of different microstructural parameters (fiber distribution, constituent properties, residual thermal stresses, etc). In the next section, the failure process is studied in detail from a micromechanical point of view.

Figure 6.3: Predicted AS4/8552 stress-strain curves for longitudinal tension under RT/DRY and HOT/WET conditions

Table 6.1: Numerically-predicted vs. experimentally-obtained elastic constants and longitudinal tension strengths for a AS4/8552 ply.

Property	RT/DRY		HOT/WET	
	Micromechanics	Marlett et al. (2011)	Micromechanics	Marlett et al. (2011)
E_1 (GPa)	136	130±3	136	130±1
X_c (MPa)	2356	1965±83	2269	1821±143

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6.1.6 Micromechanical analysis

In order to understand the failure process, a closer view to the RVE failure is required. This micromechanical study is carried out along the next paragraphs. However, before starting a detailed analysis of the results, it is worth to mention that, as it was expected, the RVE fails, due to accumulated fiber failure, through a plane perpendicular to the loading direction. In addition, since the fracture process is dominated by the fiber fracture and those are insensitive to the moisture absorption, the failure mechanism is the same for both environmental conditions (RT/DRY and HOT/WET). In view of this, the RT/DRY case will be examined in detail and the resulting conclusions can be extrapolated to the HOT/WET case.

Figure 6.4: View cut of the periodic 3D RVE FE model used in the prediction of longitudinal tension. Carbon fibers corresponding to the set of 7 breaking when maximum load is reached are highlighted (not cut).

In spite of the large number of broken fibers (26) at the end of the virtual test, a closer view to the RVE failure at the moment of failure ($\sigma = 2356 \text{ MPa}$ and $\epsilon = 1.86\%$) reveals how the number of broken fibers before reaching the maximum stress is only 7, 5 inner fibers and 2 periodic ones. This set of critical fibers, representing the 14% of the total, is shown in Figure 6.4 and it will be carefully studied along the next paragraphs.

Cutting the RVE along two longitudinal sections ($A - A$ and $B - B$) containing the critical fibers and representing their longitudinal stress distribution (σ_{11}) it can

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be seen how they are broken at the moment of failure (Figure 6.5).

Figure 6.5: Predicted longitudinal stress distribution in the AS4/8552 ply right after maximum stress ($\epsilon = 1.9\%$) under RT/DRY conditions. (a) Longitudinal section containing fibers 33, 44 and 35. (b) Longitudinal section containing fibers 50 and 33.

Isolating this group of fibers, the sequence of fracture can be studied. This process is shown in Figure 6.6, where the evolution of the longitudinal stress (σ_{11}) is plotted for each one of the critical fibers. As it can be observed, the first fracture takes place in fiber 50 ($\epsilon = 1.4 - 1.5\%$). This fiber is the only broken one up to the breakage of fibers 43 and 33 at $\epsilon = 1.7\%$. Increasing the strain up to $\epsilon = 1.86\%$ lead

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to the fracture of periodic fibers 57-7 and 55-45 together with the inner fibers 35 and 44. At this point, the RVE reaches its limit and fails. This sequence is plotted in Figure 6.7 together with the RT/DRY stress-strain curve and the RVE front view showing the relative position of the critical fibers.

Although all the broken fibers contribute to the fracture process, since the inner fibers are grouped in small clusters, a detailed analysis seems to be interesting here. In particular, two groups of fibers are studied. The first one, containing fibers 50 and 43, represents the onset of the failure process. On the other hand, the second group includes fibers 33, 44 and 35, whose failure lead to the RVE collapse. At this point it is worth to mention that two types of fiber fracture are observed in the RVE failure process: direct and induced failure. The direct failure takes place in fibers 50 and 33, whereas on fibers 43, 44 and 35, the failure is induced by the fracture of adjacent fibers.

In order to understand the fracture sequence shown in Figure 6.6 and 6.7, the longitudinal stress distribution ($\sigma(z)$) is plotted on each fiber along a longitudinal path of elements, whose nodes are the closest points between adjacent fibers. In order to provide a better comprehension, the longitudinal stresses along the fiber are normalized using the values at boundaries σ_f for each level of strain. In a similar manner, the fiber length (W) is used to normalized the distance z . The results for the different fibers are gathered in Figures 6.8, 6.9 and 6.10.

Looking at the curves gathered in Figure 6.8b, the stress concentration factor after the breakage of fiber 50, assuming that the normalized stress distribution $\sigma(z)/\sigma_f$ is a valid measurement of it, is large enough to induce fiber breakage in its neighbor, fiber 43, where a stress concentration factor of ≈ 1.3 (red line) is achieved after the breakage of fiber 50 at $\epsilon = 1.5\%$. Another important detail is that the breakage of fiber 43 is not instantaneous. Actually, the peak produced at $\epsilon = 1.5\%$ is diluted due to the presence of fiber/fiber and fiber/matrix interfaces. After the breakage of fiber 50, the stress concentration factor induces the breakage of a fiber fracture plane in fiber 43. However, at the same time, the fiber/matrix interface of fiber 43 is damaged and the stress is distributed according to the trapezoidal shape shown in Figure 6.8b (blue line). The evolution of fiber/matrix interface damage in fiber 43 is plotted in Figure 6.11 for increasing values of longitudinal strain. It is worth to mention that the extensions of the damaged zone corresponds with the regions where the stress concentration is $\sigma(z)/\sigma_f > 1$.

In the case of the ternary system composed of fibers 33, 35 and 44, the situation

Figure 6.6: Evolution of the longitudinal stress (σ_{11}) in the group of critical fibers for increasing value of longitudinal strain (ϵ_{11})

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Figure 6.7: Predicted AS4/8552 stress-strain curves for longitudinal tension under RT/DRY and HOT/WET conditions

is a little bit more complex. After the breakage of fiber 33, a stress concentration is induced in the neighboring fibers 44 and 35, triggering the onset of failure at ϵ ranging between 1.7% and 1.8% in fiber 35 (Figure 6.9) but not in fiber 44 (Figure 6.10). However, once fiber 35 collapses, an additional stress concentration is induced in fiber 44 at 1.8%. This effect is reflected in the shape of the normalized stress distribution plotted in Figure 6.10a for $\epsilon = 1.8\%$. At this point, fiber 44 is surrounded by two broken fibers. The breakage of fiber 33, induces stress concentration and delamination (see Figures 6.10b and 6.12), in a similar manner as it was shown for the binary system composed of fibers 50 and 43. However, the failure of the second one (fiber 35), is the trigger that provokes a wedge-shaped fracture starting at the fiber surface that is viewing fiber 35 (Figure 6.10a). This crack propagated from the bottom of fiber 44 to its top (viewing fiber 33). This propagation is reflected in the huge stress concentration induced in the upper part of fiber 44 (Figures 6.10b and 6.12) at ϵ ranging between 1.8% and 1.9%.

Figure 6.8: Evolution of the normalized longitudinal stress distribution along the normalized fiber distance for increasing values of longitudinal strain in the case of (a) fiber 50 and (b) fiber 43. Path front view is schematically illustrated using a red dot together with the relative fiber position.

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Figure 6.9: Evolution of the normalized longitudinal stress distribution along the normalized fiber distance for increasing values of longitudinal strain in the case of (a) fiber 33 and (b) fiber 35. Path front view is schematically illustrated using a red dot together with the relative fiber position.

Figure 6.10: Evolution of the normalized longitudinal stress distribution along the normalized fiber distance for increasing values of longitudinal strain in the case of (a) fiber 44 path 1 and (b) fiber 44 path 2. Path front view is schematically illustrated using a red dot together with the relative fiber position.

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Figure 6.11: Progressive increase of fiber/matrix interface damage in fiber 43 for increasing values of longitudinal strain

Figure 6.12: Progressive increase of fiber/matrix interface damage in fiber 44 for increasing values of longitudinal strain

6.2 Prediction of longitudinal compression

Under longitudinal compression, ply failure is triggered by the mechanism of fiber kinking, a damage mode that is strongly affected not only by the elastic properties of the fibers but also by their geometry (waviness, diameter, shape, etc.) [Budiansky and Fleck \(1994\)](#); [Berbinau et al. \(1999\)](#); [Pinho et al. \(2005\)](#); [Pimenta et al. \(2009a\)](#). In addition, void content [Bazhenov et al. \(1992\)](#); [Liebig et al. \(2016\)](#) and other imperfections (crushed fibers) arising from the manufacturing process [Soutis and D. Turkmen \(1997\)](#); [Narayanan and Schadler \(1999\)](#) can trigger the kinking.

The prediction of the longitudinal compression strength has been tackled by both analytic [Budiansky and Fleck \(1994\)](#); [Berbinau et al. \(1999\)](#); [Pimenta et al. \(2009b\)](#) and numeric approaches [Vogler and Kyriakides \(1997\)](#); [Pinho et al. \(2005\)](#); [Pimenta et al. \(2009a\)](#); [Gutkin et al. \(2010a, 2011\)](#) that simulate the complex fiber kinking phenomenon with more or less detail and accuracy. In the case of numerical simulation, models are usually based on two-dimensional micromechanical RVE's and assume perfect hexagonal fiber packing arrangements. One recent exception, allowed by the increasingly available computational resources, is the work of Bai et al. [Bai et al. \(2015\)](#). However the effect of fiber distribution, polymer plasticity and damage as well as that of the fiber/matrix debonding and friction, in the kink-band initiation and propagation is still unclear.

Computational micromechanics, based on Finite Element Analysis (FEA) offers a reliable and efficient methodology to simulate the deformation and fracture mechanisms of engineering materials. Numerical simulations of Representative Volume Elements (RVE) of the composite microstructure are able to predict ply properties, in good agreement with experimental data, e.g. [Naya et al. \(2017a\)](#); [Canal et al. \(2012b\)](#); [Herráez et al. \(2015b\)](#), providing input data for mesomechanical models that can be used in the analysis of laminates.

The aim of this section is again to develop a reliable and accurate computational approach supported by micromechanical *in situ* measurements of the material constituents to predict the longitudinal compression mechanical properties of a composite ply (stiffness and strength) under different operating environmental conditions.

In this work, following the approach of other authors [Budiansky and Fleck \(1994\)](#); [Pinho et al. \(2005\)](#); [Pimenta et al. \(2009b\)](#); [Gutkin et al. \(2010a\)](#), the initial fiber misalignment angle (ϕ_0), is assumed to control the ply failure triggering

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the fiber kinking. A three-dimensional numerical approach, that includes a coupled plasticity-damage model for the matrix and a cohesive-frictional behavior for the fiber/matrix interface, is used to investigate the influences of statistically representative microstructures and constituent mechanical properties on the longitudinal compression response of the AS4/8852 composite ply. The behavior of the micro-constituents is modeled according to the methodology described in Chapter 5.

To model this initial imperfection, the models developed in this work contain sinusoidal shaped fibers and will allow the prediction of the sensitivity function, $X_C(\phi_0)$, that represents the evolution of the longitudinal compression strength, X_C , for different values of ϕ_0 . Additionally, an initial fiber misalignment probability density, $f(\phi_0)$, is considered to achieve an objective prediction of the ply strength.

This introduction is followed by the description of the computational micromechanics framework, i.e. of the constitutive equations used to simulate matrix, fiber and interfaces, as well of the RVE generation procedure and the subsequent construction of the FE models with the specific loading conditions. Then, the results of the pure compression loading simulations performed on the particular case of the AS4/8552 UD ply are presented and correlated with reliable experimental results available in the literature [Marlett et al. \(2011\)](#). After a discussion to the methodology and results is presented, a conclusion to this work is given.

6.2.1 Initial fibre misalignment distribution

The initial fiber misalignment distribution was taken from Czabaj et al. [Czabaj et al. \(2014\)](#), who reproduced the three-dimensional real fiber distribution of a AS4/3501-6 plies laminate using high resolution images obtained by means of X-ray computed tomography. The material used by Czabaj et al. is very similar to the AS4/8552 material system used in the present work and, hence, his misalignment probability density function is used here without further modifications (see Figure 6.13). Although X-ray computed tomography is very time-consuming, this method is still faster than the one proposed by Yurgartis [Yurgartis \(1987\)](#), that consists on calculating the real (elliptic) cross section of several fibers on selected micrographs and compare these measurements against the ideal (circular) ones to obtain the rotation of the fiber section and, therefore the misalignment angle. Yurgartis' method was demonstrated to provide reproducible results [Lee and Soutis \(2007\)](#). However, it is extremely time consuming because the area of individual fibers is obtained by means of direct observation of individual micrographs captured from selected cross sections

of the composite, involving cutting, polishing and extensive observation work.

The fiber misalignment sensitivity function, $X_C(\phi_0)$, is expected to be symmetric respect to the initial fiber misalignment ϕ_0 . However, the fiber misalignment distribution function, $f(\phi_0)$, measured by Czabaj et al. [Czabaj et al. \(2014\)](#), it is clearly not. To overcome this issue, the half-function corresponding to negative misalignment (ϕ_0^-) is folded around the vertical axis (see Figure 6.13). The resulting probabilities $f(-\phi_0^-)$ are averaged with the half-function $f(\phi_0^+)$ values corresponding to positive misalignment. Then, the symmetric $\bar{f}(\phi_0)$ can be fitted by a Gaussian function, $\bar{f}_G(\phi_0)$, valid for the whole range of angles, which in turn is directly integrated to obtain the corresponding symmetric fiber misalignment cumulative density function, $F(\phi_0)$ (Figure 6.13).

Figure 6.13: Initial AS4 fiber misalignment distributions, $f(\phi_0^+)$ and $f(-\phi_0^-)$, extracted from [Czabaj et al. \(2014\)](#). Gaussian fiber misalignment probability density function, $\bar{f}_G(\phi_0)$, and cumulative density function, $F(\phi_0)$, resulting from fitting the symmetric $\bar{f}(\phi_0)$ probability function

6.2.2 Set-up and simulation of the 3D RVE models

The computational micromechanics framework is based on the analysis of a statistically representative volume element of the material (RVE) subjected to homogeneous stress states (tension, compression and shear) or temperature increments. The microstructure of the RVE of a unidirectional composite is idealized as a dispersion of parallel and circular fibers randomly dispersed in the polymer matrix. A modeling domain encompassing around 50 was found enough to capture adequately

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the essential features of the microstructure of the material [González and LLorca \(2007a\)](#). Synthetic fiber distributions statistically equivalent to the real ones are generated for the analysis using the Random Sequential Adsorption (RSA) algorithm [Segurado and LLorca \(2002\)](#). Two-dimensional periodic fiber distributions are generated and then extruded along the fiber direction to achieve the final RVE's of the unidirectional composite material. The periodic RVE's are then discretized using isoparametric linear wedge (C3D6) and brick fully integrated (C3D8) finite elements for fibers and matrix. Periodic boundary conditions are applied on opposite faces of the RVE's in three dimensions, according to the methodology developed by Segurado and LLorca [Segurado and LLorca \(2002\)](#).

In order to capture the influence of the initial fibre misalignment angle in the formation of the kink band, the total length of the RVE is set equal to one half of the wavelength associated to the sinusoidal function used to model the initial shape of the fiber (Figure 6.14). For carbon fibers, a value $W = 500\mu\text{m}$ was adopted following other authors [Gutkin et al. \(2010a, 2011\)](#). As a consequence of the large RVE size the model results in around 1.2 million DOF's, including internal variables generated to tackle the contact interactions associated to fiber/matrix interfaces. Simulations are again carried out with Abaqus/Standard [Dassault Systèmes \(2011\)](#) within the framework of the finite deformations theory taking into account residual thermal stresses generated during cool-down from the process temperature. The nonlinear problem is solved using implicit integration. The model takes over 72 hours to compute using an HPC cluster of 40 Intel Xeon E5-2670 cores and GPU acceleration.

The multi-fibre model results impractical to evaluate the sensitivity function $X_C(\phi_0)$. To improve efficiency, a simplified RVE model of a single fiber containing only ≈ 100 thousand DOF's is developed, as shown in Figure 6.15. This single-fibre RVE has the same fiber volume fraction as the multi-fibre one, but results in a reduction of the computational time to less than an hour using similar computational resources. This RVE size is convenient to perform parametric analyses and obtain an approximated $X_C(\phi_0)$ response.

Since the carbon fiber is modeled as an elastic transversely isotropic material, a local material orientation is required. To this end, the geometry of the fibers is partitioned and the resulting portions are oriented using the tangent direction corresponding to the sinusoidal-generation function at the the center of each fiber fragment. To avoid spurious element deformation modes and shear locking that

Figure 6.14: Multi-fibre FE model used in the simulation of longitudinal compression of a AS4/8552 ply. Each color represents one section. The arrows show the orientation of the fibers

would lead to numerical problems and inaccurate predictions of $X_C(\phi_0)$, it is convenient that these partitions are made using planes perpendicular to the local fiber axis instead of the global longitudinal axis. In other words, these partition planes correspond with the plane of transverse isotropy of the fibers and their orientation must change according to the sinusoidal function used to generate the model.

6.2.3 Longitudinal compression stiffness

The reliability of the modeling approach regarding elastic response is assessed by correlating its results with reliable experimental values found in the literature [Marlett et al. \(2011\)](#). The reported AS4/8552 ply longitudinal compression Young's modulus, E_{1c} has a low dependence on environment, being $E_{1c} = 114$ GPa and $E_{1c} = 116$ GPa for RT/DRY and HOT/WET conditions, respectively. Since the composite elastic response is governed by the stiffness of the carbon fibers and those are not affected by moisture absorption, these experimental observations were expected. However, reported experimental E_{1c} values are markedly lower than their

6.2. Prediction of longitudinal compression

Figure 6.15: Single-fibre FE model used in the simulation of longitudinal compression of a AS4/8552 ply (each color represents one section; arrows show the orientation of the fibers) including the geometry partition strategy used to orient the fibers and provide a regular mesh (distorted mesh resulting from using planes perpendicular to the global z axis is shown for comparison).

6. Prediction of fiber dependent ply properties

tensile counterparts, $E_{1t} \approx 136$ GPa (also fairly independent of environment conditions). In principle, since E_1 is dominated by the elastic response of the carbon fiber, the origin of this difference can be attributed either to the initial fiber misalignment ϕ_0 or to different fiber behaviors in tension and compression.

To check the first hypothesis, a parametric study relating the value of E_{1c} and ϕ_0 is conducted using the single-fiber RVE model. Results are shown in Figure 6.16 for both RT/DRY and HOT/WET conditions. It can be observed that a similar relationship between both parameters is obtained for both environment conditions when $\phi_0 < 2^\circ$. For larger values the relationship between E_{1c} and ϕ_0 diverges due to the different polymer elasto-plastic properties for RT/DRY and HOT/WET conditions.

Figure 6.16: Predicted AS4/8552 longitudinal compression modulus evolution ($E_c(\phi_0)$), for different initial fiber misalignments (ϕ_0) under RT/DRY and HOT/WET conditions. Linear and nonlinear regions are pointed out together with the corresponding fitting equations.

In order to estimate the longitudinal compression modulus of the homogenized ply, $E_{1c,ply}$, the fiber misalignment distribution of Figure 6.13 must be considered. To this end, a similar approach as the one followed by Barbero [Barbero \(1998\)](#) to determine the ply compression strength, $X_{c,ply}$, is adopted here. This method states that the applied load function, $\tilde{\sigma} = X_c(\phi_0)F(\phi_0)$, represents the effective stress on fibers that remain unbroken or un-kinked. This load function has a maximum

6.2. Prediction of longitudinal compression

that corresponds to the ply compression strength $X_{c,ply}$. Therefore, substituting the misalignment sensitivity function $X_c(\phi_0)$ by the $E_{1c}(\phi_0)$ relationship of Figure 6.16, for each of the environment conditions, $E_{1c,ply}$ is predicted as the maximum of the curve $E_{1c}(\phi_0)F(\phi_0)$, hence obtaining $E_{1c,ply} = 130$ GPa and $E_{1c,ply} = 129$ GPa, for RT/DRY and HOT/WET conditions, respectively. However, these values still represent an important over-prediction of the ones obtained experimentally Marlett et al. (2011). Therefore, it can be concluded that ϕ_0 has a minor influence on the longitudinal compression modulus of the composite.

In view of this, and since the fiber properties used in the computational model were obtained under tensile loads, the difference between the ply longitudinal tension and compression moduli seems to derive mostly from the fiber elastic response rather than from geometrical imperfections. This idea has been pointed out by several authors, e.g. Oya and Johnson (2001); Mujika et al. (2006), who stated that the longitudinal modulus of carbon fibers under tension (E_{ft}) and compression (E_{fc}) can be quite different. In particular, Mujika et al. (2006) has shown that $E_{fc}/E_{ft} = 0.88$ for AS4 carbon fibers. The origin of this phenomenon is still not clear, hence this assumption needs to be considered carefully. High resolution images of PAN-based carbon fiber longitudinal and transverse sections have shown that there is a complex interlinking of layer planes both longitudinally and laterally Bennett et al. (1983). The presence of misoriented crystallites, with respect to the fiber axis, could explain the lower value of the compression modulus compared to the tension one. However, more research work is required to understand the mechanical response of the carbon fiber inner structures.

Applying the correction factor proposed by Mujika et al. (2006) to the modulus determined using Barbero's method Barbero (1998) leads to set of ply compression modulus that are in good agreement with the ones experimentally obtained by Marlett et al. (2011), as presented in Table 6.2.

6.2.4 Longitudinal compression strength

Although, the computation of the misalignment sensitivity function $X_c(\phi_0)$ is not practical with the multi-fibre model (Figure 6.17), this RVE is able to capture the initiation of the fiber kinking damage process as well as showing some relevant post load-peak features. Following the notation used by Pimenta et al. (Pimenta et al. (2009a)), the fiber rotation angle (α) and band width (w) can be pointed out representing the RVE in the deformed configuration for increasing strains, as it is

shown in Figure 6.17c for an initial fiber misalignment $\phi_0 = 3^\circ$.

These results, $\alpha \approx 14^\circ$ and $w \approx 120 \mu\text{m}$, are in good agreement with experimental data Gutkin et al. (2010b); Jelf and Fleck (1994); Pinho et al. (2006b), as well as with other numerical results already published Bai et al. (2015); Pimenta et al. (2009b); Gutkin et al. (2010a) for similar values of applied strain. In particular, Gutkin et al. Gutkin et al. (2010b) pointed out that the kink band width is in the order of 10-30 fiber diameters. Nevertheless, it is worth to mention that the RVE approach is not suited to capture the propagation of damage but its initiation. In fact, if periodic boundary conditions are used, the RVE corresponds to a UD ply region that is far away from the borders, which means that the orientation of the kink-band angle, β , is zero as it shown shown, for example, by Gutkin et al. Gutkin et al. (2010a). Fortunately, imposing periodic boundary conditions on lateral surfaces will not influence the predictions of strength since the kink-band angle only starts increasing after the peak load Pimenta et al. (2009a).

On the other hand, unlike the multi-fibre RVE, the efficient 3D single-fiber RVE model cannot capture these kink band "propagation" features but its predictions of the compression elastic modulus and of the kink load (X_c) is the same obtained by the multi-fibre RVE for the same initial fiber misalignment (see Figure 6.17a). For the particular case of our AS4/8552 coupons, the interface is weaker than the matrix and, hence, the ply strength is governed by the fiber volume fraction and the initial misalignment instead of by the fiber distribution (inter-fiber spacing). If the interface is stronger than the matrix, the fiber distribution will influence (theoretically) where and when the onset of plastic deformation takes place in the matrix. In view of this, the efficient 3D single-fiber RVE can be used to perform parametric analyses and obtain $X_c(\phi_0)$ corresponding to these AS4/8552 coupons.

Modeling the fiber behavior as linear-elastic leads to unrealistic predictions of X_c for small values of the initial misalignment angle (roughly $0^\circ < \phi_0 < 1.0^\circ$), since under such conditions fiber crushing is bound to occur at lower loads than fiber kinking Gutkin et al. (2010a). To account for this phenomenon in an approximated way, an isotropic continuum damage model available in the library of ABAQUS Dassault Systèmes (2011) is used. The authors infer that the fiber longitudinal compression modulus should be lower than the tensile one due to the morphology of the carbon fiber cross section Bennett et al. (1983). However, the necessary fiber characterization results are not available. Hence, values found by Herráez et al. Herráez et al. (2016) for longitudinal fiber tension are used instead.

6.2. Prediction of longitudinal compression

Figure 6.17: Predicted kink-band development in AS4/8552 under RT/DRY conditions and pure longitudinal compression: a) Plastic equivalent strain (PEEQ) at two load states: A - kink-band initiation corresponding to peak load; and B - fully developed kink-band; b) Stress-strain curve - Comparison between complex and simplified RVE for the same initial fiber misalignment, $\phi_0 = 3^\circ$; c) Plastic equivalent strain (PEEQ) evolution with prediction of kink-band width and fiber angle α .

Figure 6.18: Predicted stress-strain curve for AS4/8552 ply under pure longitudinal compression. Comparison between complex and simplified RVE for the same initial fiber misalignment, $\phi_0 = 3^\circ$

Figure 6.19: Predicted plastic strain evolution of AS4/8552 ply for longitudinal compression under RT/DRY conditions. Plotting the deformed configuration for different strains points out kink band initiation and propagation

6.2. Prediction of longitudinal compression

By using this approach, the sensitivity function $X_c(\phi_0)$ is finally calculated for the different initial fiber misalignment values and both RT/DRY and HOT/WET environmental conditions. The numerical results are gathered in Figure 6.20a and compared to the analytically solutions implemented in LaRC04 [Pinho et al. \(2005\)](#) and developed by Pimenta et al. [Pimenta et al. \(2009b\)](#). Remarkable correlation is obtained with LaRC04. The model of Pimenta et al. tends to overpredict X_c due to the assumption of an elastic-perfectly plastic matrix behavior. Both LaRC04 and present numerical approach adopt more realistic matrix plastic hardening response. Under RT/DRY conditions, the sensitivity function presents two different regions. Up to $\phi_0 = 1.0^\circ$, the failure mechanism is fiber crushing and the fracture process is governed by the fiber compression strength. For $\phi_0 > 1.0^\circ$, the initial fiber misalignment and matrix plasticity controls the failure of the RVE, and therefore the failure occurs by kink band formation. In the case of HOT/WET conditions, the effect of fiber crushing is almost negligible ($\phi_0 < 0.5^\circ$). Nevertheless, under both conditions, the predicted values of $X_c(\phi_0)$ are clearly lower than the ones predicted by the analytic solution. This is due to the presence damageable fiber/matrix interface, whose failure triggers the onset of matrix plastic deformation, but also due to the matrix damage and carbon fiber anisotropy.

The actual ply compression strength $X_{c,ply}$, is predicted using the method developed by Barbero [Barbero \(1998\)](#) introduced above. The nominal compression strengths of the composite ply are given by their maximum values of the applied load function, $\tilde{\sigma} = X_c(\phi_0)F(\phi_0)$. The predicted load functions $\tilde{\sigma}(\phi_0)$ for RT/DRY and HOT/WET conditions are shown in Figure 6.21. As presented in Table 6.2, the numerical results are in excellent agreement with experimentally-obtained values found in the literature [Marlett et al. \(2011\)](#) for both environmental conditions.

Table 6.2: Numerically-predicted vs. experimentally-obtained elastic constants and longitudinal compression strengths for a AS4/8552 ply.

RT/DRY		HOT/WET	
Property	Micromechanics Marlett et al. (2011)	Micromechanics Marlett et al. (2011)	
E_1 (GPa)	114	113.5±2	113
X_c (MPa)	1356	1441±43	1064
			1065±30

Figure 6.20: a) Predicted AS4/8552 ply compression strength evolution ($X_c(\phi_0)$), for different initial fiber misalignment (ϕ_0) under RT/DRY and HOT/WET conditions. Comparison between present FE simulations and analytical solutions implemented in LaRC04 [Pinho et al. \(2005\)](#) and developed by Pimenta et al. [Pimenta et al. \(2009b\)](#). Schematic representation of the matrix elasto-plastic response adopted by both Pimenta's and LaRC analytical solutions is shown in (b) and (c) respectively.

6.2.5 Fiber kinking micromechanisms

With the purpose of studying the fiber kinking micromechanisms, in combination with the influence of environmental conditions, the single-fibre configurations corresponding to the critical values of the initial fiber misalignment angle for RT/DRY and HOT/WET conditions, $\phi_0 = 1.31^\circ$ and $\phi_0 = 1.14^\circ$ respectively, are analyzed here in great detail (see Figure 6.22a).

Using the HOT/WET case as reference (similar mechanisms are captured for the RT/DRY condition), evolution of the interface scalar damage variable is plotted for progressive values of applied strain in Figure 6.22c. Paying attention to the longitudinal and cross paths shown on the top of Figure 6.22c, the evolution of delamination along the longitudinal and transverse direction (red triangles and black diamonds, respectively) and the matrix plasticity along the cross section, represented using the blue squares, is monitored and shown in Figure 6.22b. It can be observed that the kinking process is triggered by failure of the interface that initi-

6.2. Prediction of longitudinal compression

Figure 6.21: Barbero's load function $\tilde{\sigma}(\phi_0)$ predicted for AS4/8552 under RT/DRY and HOT/WET conditions.

ates at the intersections of the fiber surface with the misalignment plane. In these stages ($0.008 < \epsilon < 0.01$), interface damage propagates along both longitudinal and circumferential paths, reducing the load transferred between the fiber and the matrix. This process, which is controlled by the interface energy release rate, continues until the onset of matrix damage ($\epsilon \approx 0.01$). At this point, longitudinal delamination is arrested and transverse delamination increases rapidly until almost the whole cross section is damaged ($> 90\%$ of the total circumference perimeter) making the RVE unstable ($0.0105 < \epsilon < 0.0106$). This point corresponds to the maximum load supported by the RVE. During the post-peak ($\epsilon > 0.0106$), the longitudinal delamination grows again and matrix undergoes severe plastic deformation.

To highlight the role of the fiber/matrix interface in the fiber kinking process, the RVE analyses are repeated using perfect interfaces. The stress-strain results corresponding to realistic and perfect interfaces are compared in Figure 6.22a for both environmental conditions. It can be concluded that interface failure triggers the collapse of the RVE, leading to important reductions of the maximum applied loads in comparison with analyses considering perfect interfaces. Such influence of the interface was reported by Madhukar et al. [Madhukar and Drzal \(1991\)](#), who studied the effect of interface strength in the longitudinal compression strength and as well as in the type of failure mechanism. The load drop (22%) is specially important

6. Prediction of fiber dependent ply properties

in the HOT/WET case because of the lower interface strength measured in this condition (see Table 5.3).

It must be pointed that this analysis is only valid for the particular case of the AS4/8552 composite. In another material, with different matrix, fiber/matrix interface properties and fiber volume fraction, a different interaction of failure mechanisms might be observed.

Figure 6.22: a) Predicted AS4/8552 ply compression strength for critical initial fiber misalignment (ϕ_0) under RT/DRY and HOT/WET conditions. Comparison between realistic and perfect interface cases; b) Evolution of interface damage (along fiber direction and transverse sections perimeter) and matrix plasticity along transverse section perimeter for the case of real interface under HOT/WET conditions; c) Progressive increase of interface damage with applied compression load for the case of real interface under HOT/WET conditions.

6.3 Concluding remarks

Continuing the work presented in Chapter 5, a coupled experimental-computational micromechanics framework is proposed to simulate the fiber dependent properties of unidirectional composite plies allowing the virtual investigation of environmental effects. The approach is again based on the *in-situ* experimental characterization of the microconstituents. Using only material constituent properties, the models developed are able to reproduce in detail the experimentally observed ply behavior although at a high computational cost.

Trying to improve the computational efficiency, simplified models are also proposed that provide still accurate results for the most important ply properties. In the case of longitudinal tension, matrix plasticity is removed, since it does not affect the ply strength value (at least for the particular case of our AS4/8552 coupons). However, calculations are still highly non-linear, requiring an important amount of hardware. Actually the number of fibers and fracture planes used in the 3D RVE FE model was increased as much as possible according to system memory limitations.

In the case of longitudinal compression, simplified models (single fiber RVE) offered an important saving of computation time. This advantage was used to assess the influence of the initial fiber misalignment in the final ply strength. It is worth to mention that it was assumed that the major feature affecting fiber kinking is the initial fiber misalignment generated during composite manufacturing. Other manufacturing defects have been demonstrated by other authors to have effects on the compression strength, but they were considered here to be minor and not analyzed. Moreover, The experimental measurements of this property to which the present results are correlated are likely to be affected by the experimental set-up, by the sample size and geometry, among other factors. In view of this, the present virtual test approach can be understood to provide the upper limits of the ranges of variability of the measured properties.

Under longitudinal tension, the process is dominated by the presence and distribution of volumetric defects. The fracture process at the microlevel has a strong local component related to how the stress is redistributed after the breakage of a ceramic fiber. Since the fiber fracture process has stochastic character and is highly influenced by the number of fracture planes and fibers present in the RVE, simplified models like the single fiber RVE used in longitudinal compression are not accurate enough. In addition, although Weibull distribution is suitable to capture the fiber

6. Prediction of fiber dependent ply properties

strength distribution, a GLS model ([Curtin, 1991](#)) does not provide accurate results at the micro scale, because it puts aside relevant local information, such as the stress concentration and interface debonding, and, hence, it trends to overestimate the longitudinal ply strength.

The role of the fiber/matrix interface was also investigated by micromechanical analysis. In the case of longitudinal tension, debonding started after the first fiber breakage explaining the origin of the small non-linear region present in the stress-strain curve before reaching the maximum stress. It also explains the small decrease of tension strength for HOT/WET conditions. Nevertheless, it has a minor influence compared to the fiber strength distribution, that is controlled by the Weibull parameters and the number of fibers present in the RVE.

On the other hand, fiber/matrix interface strength plays a major role in fiber kinking, triggering the RVE failure. Important strength reduction between a composite ply with perfect and real interface is found. In view of this, developing new analytical models considering this micromechanical parameter are desired.

In summary, this chapter, in a similar fashion to the previous one, proposes a virtual ply characterization methodology based on reliable properties of the micro-constituents.

Conclusions and future work

7

7.1 Conclusions

According to the author, the main conclusion or, in other words, the main achievement of this thesis is the development of a coupled experimental-computational strategy based on a micromechanics framework that is able to predict the mechanical response of a UD FRP ply under different loading conditions and environments. The proposed methodology has been tested using a carbon/epoxy unidirectional laminate of aeronautical grade made of AS4/8552 pre-impregnated (pre-preg) laminae under two different environmental conditions (RT/DRY and HOT/WET).

The predicted ply properties, classified in matrix-dependent (transverse tension-compression and in-plane shear) and fiber-dependent properties (longitudinal tension-compression) are in good agreement with the values reported in the literature ([Marett et al., 2011](#)). In addition, the numerical models reproduce the elasto-plastic ply stress-strain behavior and fracture mechanisms observed experimentally [Canal et al. \(2009\)](#); [Totry et al. \(2010\)](#); [González and LLorca \(2007a\)](#), both for uniaxial and biaxial stress states, suggesting that virtual ply characterization tests, based on reliable properties of the microconstituents, can replace the physical experiments, at least for material screening purposes. Actually, the resulting properties, obtained by this bottom-up multiscale approach have been successfully used to predict the response of composite laminates at the mesoscale level [Lopes et al. \(2016\)](#); [Falcó et al. \(2017\)](#).

The work carried out in this thesis can be classified in two different but complementary topics, experimental and computational micromechanics, and so the conclusions will be.

7.1. Conclusions

7.1.1 Experimental micromechanics

Since the work carried out in thesis was framed into a bottom-up multiscale strategy, one of the main goals was the characterization of the composite constituents under different environmental conditions by means of micromechanical testing and, hence, avoiding any type of macroscopic test dependency. In addition, since the access to separate constituents is often not possible, the micromechanical tests should be able to be carried out using the composite. To this end, the elasto-plastic behavior of a polymer matrix has been evaluated by means of a novel methodology combining two different *in-situ* micromechanical techniques, namely the instrumented nanoindentation and the micropillar compression, performed directly in the composite cross section.

Although instrumented nanoindentation offers the best predictions of the polymer elastic modulus, micropillar compression shown a set of very interesting features that pointed out potential applications. Unlike instrumented nanoindentation, micropillar compression tests provides the whole polymer stress-strain curve. In addition, the uniaxial stress-state achieved during the pillar compression gives a direct measurement of the mechanical properties. This easy interpretation of the measurements constitutes a great advantage compared to the instrumented nanoindentation, where the determination of the contact area require the use of computational models relating the different mechanical parameters (Rodríguez, 2012). Nevertheless, the micropillar compression technique is not exempt of problems when applied to polymeric materials. Both the advantages and drawbacks of each technique have been investigated in this work (see Chapter 5).

It is worth to mention that the same set of properties, including those measured by means of micromechanical tests, have been used to predict the UD ply mechanical response regardless of the loading conditions. The same constitutive equations calibrated with the same set of properties have been used to predict both matrix and fiber-dependent ply properties without the need of any non-physical fitting parameter. From the author's point of view, this is one of the main achievement of the bottom-up multiscale approach carried out in this work.

7.1.2 Computational micromechanics

The microscopic failure mechanisms have been captured for both matrix and fiber dependent mechanical properties. Under tensile loading, failure took place along a

plane perpendicular to the loading direction. In the transverse case, fiber/matrix interface debonding triggered the material softening, whereas under the longitudinal loading, fiber failure controlled the composite failure process. As a general conclusion, the transverse and in-lane shear behavior is controlled by matrix plasticity, whereas longitudinal tension and compression is governed by fiber defects (volumetric flaws and initial misalignment) and, in both cases, fiber/matrix interface plays an important role in the failure process.

Both fiber and matrix dependent ply properties have been determined using 3D RVE FE models that take into account the real fibre distribution (fibre packing, clusters, matrix rich regions, etc. can be taken into account) and calibrated using the properties measured by micromechanical testing as much as possible. This type of approach provides information regarding the importance of each constituent in the different failure modes and is able to reproduce the stress-strain curves (without fitting parameters), as well as the deformation and fracture mechanisms experimentally observed representing a bottom-up multiscale approach.

Certainly, full-scale models are able to reproduce in detail the experimentally observed ply behavior, although at a high computational cost. In view of this, another important conclusion concerns the usage of simplified models to reduce the required computational resources. This is for example the case of removing matrix plasticity from longitudinal tension models or using single-fiber RVEs to predict the tensile failure and the fiber kinking respectively. These simplified models provide a affordable approach, from a computational point of view, for future parametric studies aimed to evaluate the influence of constituent properties in the composite material mechanical response.

In summary, this thesis proposes a virtual ply characterization methodology based on reliable properties of the microconstituents, aimed to replace the physical experiments, at least for material screening purposes. These virtual tests provide full control of the composite microstructure and constituent properties, allowing microstructural optimization to be performed in the future. Moreover, the computational approach allows the simulation of complex stress states, not possible to be applied experimentally. Thanks to this multiscale bottom-up approach, the importance of some micromechanical parameters, such as fiber distribution, cohesive-frictional fiber/matrix interface properties, fiber behavior under tension and compression can now be assessed representing an interesting research line to be explored by future investigations.

7.2 Future work

7.2.1 Experimental micromechanics

In this thesis, Focused Ion Beam (FIB) has been pointed out as a very useful tool for micromechanical testing (micropillar milling) applied to composite materials. Actually, the number of potential applications is huge. In view of this, thinking about new types of tests aimed to obtain the mechanical response of the different constituents at the micro level would be of great interest. Material testing at such small scales can provide useful information to be used for microstructure optimization.

Taking advantage of the FIB capabilities, new fibers and matrix *ad-hoc* specimens can be manufactured opening a new set of possibilities. As an example, new ways of measuring tensile behavior and interface strength of composite materials are nowadays an active research line ([Barber et al. \(2015\)](#); [Shih et al. \(2013\)](#)).

On the other hand, the assumption of a constant polymer internal friction angle regardless of temperature and water absorption, true for certain epoxy resins, should be verified for other type of polymers. Among the possible testing methods, the combination of instrumented nanoindentation and micropillar compression constitutes a promising alternative. Actually, the combination of both techniques can be used to determine the material elasto-plastic response at different temperatures and strain rates, following the preliminary work shown in Chapter 4.

Regarding the AS4/8552 fiber/matrix interface, material hydrothermal aging dropped its strength, unlike the results shown by [Rodríguez \(2012\)](#) for a similar material system. In addition, the push-in tests used to measure it were carried out at room temperature (due to hardware limitations), which means that the simultaneous action of water absorption and temperature increase was not evaluated. Repeating the tests at high temperature for both dry and wet samples and validating the extrapolation used in Chapter 4 is a clear pending task. In addition, the role of friction proved to be very relevant when transverse compression stress is present. To this end, new tests aimed to measure the friction coefficient at different environmental conditions must be developed (see [Shih et al. \(2013\)](#)).

Another important issue that is still unsolved is the development of efficient experimental methods to measure the fracture toughness of the different constituents at the microscale. Specially relevant is the case of the fiber/matrix interface, where matrix plasticity, friction and debonding are acting at once. Developing reliable

tests, valid for different environmental conditions, constitutes a tremendous challenge that must be tackled in the near future in bottom-up multiscale approaches.

Finally, studying the distribution of volumetric defects, such as pores and broken fibers, as well as the shape of the reinforcements is desired in order to improve the prediction of the composite mechanical response. This issue is specially relevant for fiber-dependent ply properties, where defects govern the failure process. To this end, X-ray computed tomography represents a promising alternative (Requena et al. (2009); Czabaj et al. (2014)). Nevertheless, the improvement of the experimental procedures and their current resolution is still a pending task.

7.2.2 Computational micromechanics

As it was pointed out in the conclusions, full-scale models are really computationally exigent, specially for fiber dependent ply properties. Improving the numerical models, making them more robust and efficient is mandatory, specially if new biaxial and triaxial stress states want to be predicted. This seems to be the next step to take advantage of 3D RVE FE models. To this end, X-FEM methods to deal with matrix fracture and improved cohesive elements (including the effect of friction under compression) could provide a computationally efficient solution.

Although the numerical predictions obtained for the AS4/8552 are in good agreement with the values reported in the literature, the strategy developed in this thesis must be validated not only for new material systems but for more complex models in the mesoscale level, as it is expected for a multiscale approach.

However, moving towards the top of the multiscale pyramid rapidly increases the problem difficulty. In order to get accurate predictions, evaluating the number and type of defects and, hence, allowing the simulation of stochastic phenomena is a major goal. Trying to incorporate the real manufacturing conditions (thermal gradients, dependency of properties with temperature, geometrical restrictions, etc.) to the micromechanical models will allow the development of defects in a natural way. Although it represents an important challenge, this is a crucial step to reproduce the transition from the microlevel to larger scales in an accurate way.

Finally, in line with the bottom-up multiscale strategy developed along this thesis, the development of new failure criteria influenced by relevant micromechanical information, such as fiber distribution, cohesive-frictional fiber/matrix interface properties, fiber behavior under tension and compression, etc., aimed to improve

7.2. Future work

the state-of-the-art failure criteria currently available ([Dávila et al. \(2005\)](#); [Puck and Mannigel \(2007\)](#); [Catalanotti et al. \(2014\)](#)).

Summary of FRP failure criteria

Using the nomenclature shown in Chapter 2 (Figure 1.5), the following equations can be used to predict Puck's failure envelope for matrix dependent properties (inter fiber failure [Puck et al. \(2002\)](#)).

Mode A: $\sigma_2 \geq 0$

$$f_E = \sqrt{\left[\left(\frac{1}{R_{\perp}^t} - \frac{p_{\perp\parallel}^t}{R_{\perp\parallel}} \right) \sigma_2 \right]^2 + \left(\frac{\tau_{21}}{R_{\perp\parallel}} \right)^2 + \frac{p_{\perp\parallel}^t}{R_{\perp\parallel}} \sigma_2} \quad (\text{A.1})$$

Mode B: $\sigma_2 < 0$ and $\left| \frac{\sigma_2}{\tau_{21}} \right| \leq \left| \frac{R_{\perp\perp}^A}{\tau_{21,c}} \right|$

$$f_E = \sqrt{\left(\frac{\tau_{21}}{R_{\perp\parallel}} \right)^2 + \left(\frac{p_{\perp\parallel}^c}{R_{\perp\parallel}} \sigma_2 \right)^2} + \frac{p_{\perp\parallel}^c}{R_{\perp\parallel}} \sigma_2 \quad (\text{A.2})$$

Mode C: $\sigma_2 < 0$ and $\left| \frac{\sigma_2}{\tau_{21}} \right| \geq \left| \frac{R_{\perp\perp}^A}{\tau_{21,c}} \right|$

$$f_E = \left[\left(\frac{\tau_{21}}{2(1 + p_{\perp\perp}) R_{\perp\parallel}} \right)^2 + \left(\frac{\sigma_2}{R_{\perp}^c} \right)^2 \right] \frac{R_{\perp}^c}{-\sigma_2} \quad (\text{A.3})$$

, where $R_{\perp}^t$, $R_{\perp\parallel}$ and $R_{\perp}^c$ corresponds to Y_T , S_L and Y_C , receptively. On the other hand,

$$\tau_{21,c} = R_{\perp\parallel} \sqrt{1 + 2p_{\perp\perp}^c} \quad (\text{A.4})$$

and

$$R_{\perp\perp}^A = \frac{R_{\perp}^c}{2(1 + p_{\perp\perp}^c)} \quad (\text{A.5})$$

The failure envelope is calculated giving values to σ_2 and solving for τ_{21} or vice-versa. The Newton-Raphson algorithm is used to solve the resulting non-linear problem.

The following equations can be used to predict the LaRC failure envelope for fiber dependent properties (longitudinal tension and compression).

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