EFFECT OF THE INTERPHASIAL DEBONDING ON THE ELASTIC BEHAVIOR OF UNIDIRECTIONAL GLASS-FIBER/EPOXY COMPOSITES

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Abstract
Due to the presence of the sizing substance during the fiber surface treatment, an interphase area is often present at the vicinity of the reinforcements. Using a FEA homogenization technique, this study presents the influence of the interphasial debonding on UD composites. Numerical models using ABAQUS™ are developed in order to predict the mechanical behavior of a unidirectional composite (E-glass fibers/epoxy) under monotonic transverse traction. A Representative Volume Element description with a random distribution of the fibers is used. Kinematic Uniform Boundary Conditions are considered and the hypothesis of a linear elastic matrix and fibers is made. The interphases are described using cohesive elements and the effect of the interphase modulus and its thickness on the global behavior of the composite is discussed. Finally, a numerical/experimental comparison is made to validate the model. The thickness and the modulus of the interphase have been determined by experimental measurements based on Atomic Force Microscopy and introduced in the numerical simulation. The obtained results are in good agreement with the experimental values previously measured by tensile test on macroscopic samples. The fact of considering the interphase improves the accuracy of the prediction of the composite's mechanical elastic behavior.

1. Introduction

Glass fibers are the most employed reinforces to manufacture composites excluding reinforced rubber. Fibers are stretched up to a diameter of 5 µm to 15µm [1]. These fibers have a brittle elastic behavior and are susceptible to abrasion damage. This is the reason why they are coated by a resin, which ensures fibers protection. This chemical substance is called sizing (Fig. 1 [2]) and contains an adhesion promoter (usually an organosilane compound as coupling agent), a film former along with a suitable emulsifier and a lubricant [3]. During composites fabrication, a chemical reaction occurs modifying the matrix's network at the vicinity of the reinforcement. A new region of finite dimensions is created between the matrix and the coated fibers; this so-called interphase has mechanical properties varying from those of the bulk phases.

The qualities of a composite material and especially its mechanical performances are directly linked to the interphase's quality [4]–[6]. The formation of both a softer and a harder interphase is possible, depending on the combination of reinforcement, matrix and coupling agent applied. Development of a strong fiber/matrix interphase, or lack thereof, has a significant effect on the failure mode and fracture surfaces of composites [7]. Moreover, the composite's rupture (frequently decohesion [8]) under transverse loads begins at the fiber/interphase interface and thus its properties greatly affect the transverse strength of the material [9], [10]. Therefore, the knowledge of the influence of this sizing on the interphase's formation represents a precious insight for industrial matters.

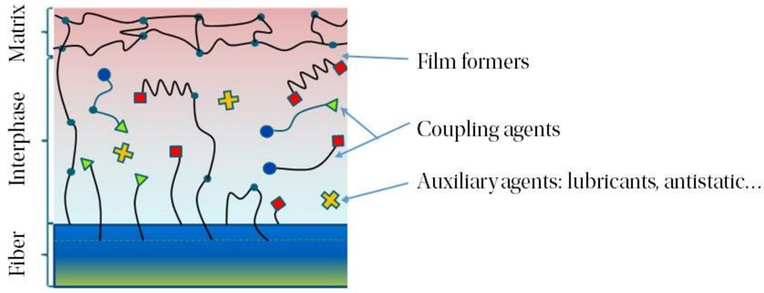


Figure 1. Schematic representation of the interphase due to sizing treatment of the fiber.

Although the interphase may seem small (in the order of nm), its mechanical properties can significantly affect the overall composite behavior [11]. Therefore, to enhance performance in composites materials, adhesion between matrix and reinforcements should be of primary interest and thus it is crucial to understand the effect of the interphase debonding on the overall behavior of the composite. Any changes in the parameters of the interphase or any of the constituents of the composite implies additional measurements to determine the overall behavior of the structure or macroscopic sample. This may become time consuming and cost prohibitive [12]. Therefore, numerical analysis and specially micromechanical techniques are essential to predict the composite behavior including the interphase. The upward trend of the evaluation and characterization of microscale material structure/property has led to an increased understanding of the effect of the interphase region on the performance of composite materials [13]; however, our understanding of this interphase is far for being complete. Different techniques could be used to highlight this interphase, such as the Scanning Electron Microscope (SEM), the Transmission Electron Microscope (TEM) or few spectroscopic analysis: Fourier Transform Infrared (FTIR) spectroscopy, Time-of-Flight Secondary Ion Mass Spectrometry (ToF-SIMS), Auger Electron Spectroscopy (AES) and X-ray Photoelectron Spectroscopy (XPS). As the results obtained from these methods reveal trivial information concerning the mobility of the macro-molecular chains, further analyses need to be inquired using specific techniques as Atomic Force Microscopy (AFM) [14] or Dynamic Mechanical Analysis (DMA) for example, which intensify the details about the local response [15] giving information related to the mobility of the polymer chain segments to improve the characterization of interphase's properties [16], [17].

The aim of the research is to accurately simulate the macroscopic behavior of an UD glass-fiber/epoxy composite through a multi-scale approach considering interphases described as cohesive elements and based on the results from previous work [18]. As mentioned before, previous literature neglect this feature in composites, thus we intend to prove its importance in numerical analysis. The behavior of the matrix and fibers have been simplified as to linear elastic constitutive materials, the effect of the plasticity of the matrix was considered in previous works [19] concluding in the non-significance of this parameter, however this phenomenon is studied to confirm its influence on the macroscopic response of the composite when damage mechanisms are taken into account [9], [20]. Regarding the composite treated in the present paper, as it is a transversely isotropic material and considering a plane strain state, each constituent is equivalent isotropic. Considering these hypothesis, our aim is to illustrate the sensitivity of the mechanical response to the existence of an interphase discretized by *"solid homogeneous"* or *"cohesive"* elements. Hence, the thickness and the elastic modulus of the interphase are determined by experimental measurements based on AFM, these values are injected on a numerical RVE with random distributions of the fibers. The geometry of this micro-model has been generated using an ABAQUS™ plug-in developed by the first author. An FEA homogenization technique is used considering Kinematic Uniform Boundary Conditions and linear elastic constitutive materials. Furthermore, we expose an analysis pointing out the influence of the cohesive element parameters (used for the interphase) on the overall mechanical properties of composite. A numerical/experimental comparison based on a monotonic traction submitted in the composites transverse section is made, in order to validate the model. The fact of considering the interphase improves the accuracy of the prediction of the composite's mechanical behavior. Moreover, the model could be used to predict the interphase properties (thickness, Young's modulus and Poisson's ratio) of a composite knowing the macroscopic stress–strain response or the elastic modulus of the overall composite.

2. Materials and methods

In this section, the composite has been mechanically characterized from the submicron scale to the macroscopic scale. A composite plate based on epoxy matrix with glass fibers was prepared by filament winding process. The thermosetting matrix is a DGEBA-based epoxy resin reinforced by E-glass fibers treated with a commercial sizing. The porosity resulting from the process is lower than 1% volume fraction for a fiber content around 54% volume fraction.

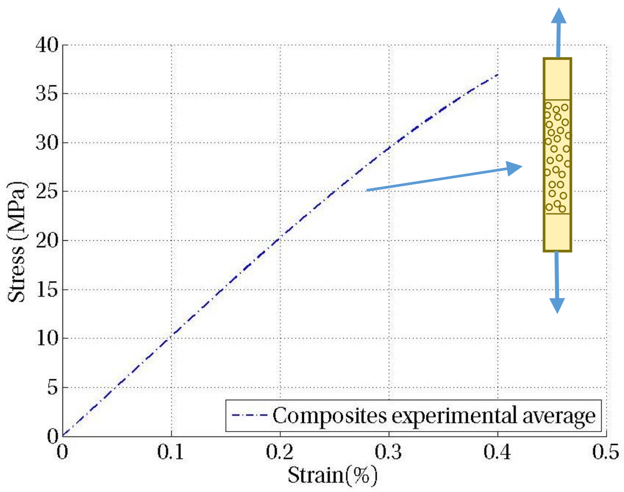


Figure 2. Composite mechanical behavior, transverse traction.

Tensile tests are realized to characterize the mechanical behavior of the samples. They were all performed on a MTS DY35 machine equipped with a 20kN force sensor. The UD composite samples are placed in the transverse direction to highlight the fiber/matrix interface [15]. The tensile test conditions and the dimensions of the test specimens are defined by the standard NF EN ISO 527–5. The crosshead speed used was 1 mm/min. Parallelepiped samples of 250×25×2mm3 with glass epoxy end tabs were prepared. The experimental average result show a brittle behavior for the UD composite in transverse direction (Fig. 2). This curve was obtained by averaging five repeatable test and the average value of Young modulus calculated from is equal to 10GPa. This result will be compared to the results from the numerical models.

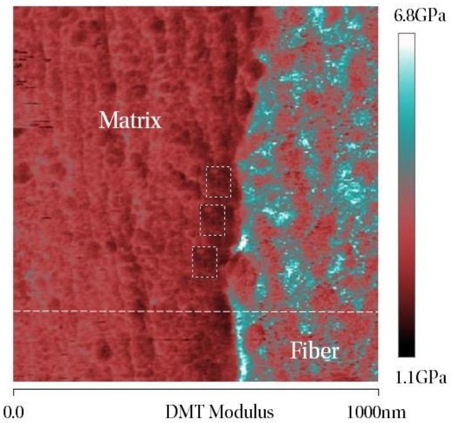
Table 1 resumes the material properties used in this work as input data for numerical simulations. As mentioned before, interphase and matrix elastic modulus are obtained by experimental measurements. Glass-fiber properties were extracted from commercial data sheets. The Poisson's ratio of the matrix is acquired from literature [21] and the interphase Poisson's ratio is assumed equal due to our incapability of measuring it experimentally. However, a numerical sensitivity analysis of the interphase Poisson's ratio is proposed below in Section 3.3.

Table 1. Constituent properties used in the numerical model.

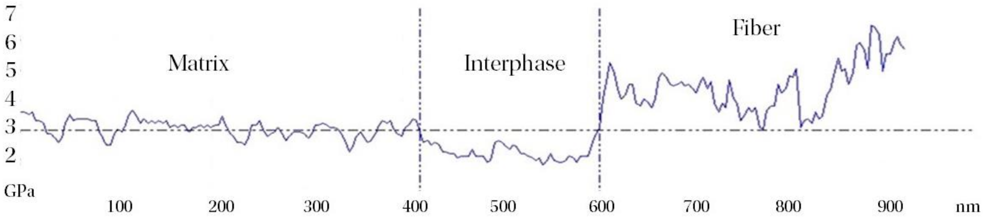
Constituent	*Young's modulus* (GPa)	*Poisson's ratio* (GPa)
Matrix	2.7 ± 0.3	0.4
Fibers	80	0.25
Interphase	1.7 ± 0.2	0.4

2.1. Atomic Force Microscopy

This technique is used to characterize mechanical properties at the surface of the samples and more especially near the fiber, in interphase areas. The nanomechanical mode PEAKFORCE QNMTM can perform force curves at a frequency of 2 kHz while scanning the surface of the samples. The forces applied remains very low (25 nN) to minimize the impact of each measurement on surface properties and improve lateral resolution. Force curves are analyzed in real time to determine the local elastic property at the nanoscale from the DMT model (Derjaguin Muller Toporov) [22]. Height scans as well as scans in modulus or adhesion can hence be obtained simultaneously. The analyses are performed on a BRUKER V9 MULTIMODE2-U equipped with the NanoScope MM-PFQNM. The cantilevers on composite samples are BRUKER RTESPA type which stiffness is around 40 N/m. They are calibrated in stiffness on a sapphire specimen and thermal tune integrated procedure. Tip radius are then determined from scans on polystyrene reference samples with known modulus.



(a) Local modulus mesurements.



(b) Evolution of Young's modulus.

Figure 3. Composite mechanical behavior, transverse traction.

The samples were cut perpendicular to the fibers and polished down to 0.05 μm with an alumina suspension for less than one minute to avoid fiber degradation. The polishing residues are removed after rinsing with distilled water and blowing with dry air. The modulus measurements on the composites are obtained from a constant force around 25 nN and the modulus profiles are obtained on scans between 300 nm and 1 μm length. We must highlight that the fiber modulus cannot be measured with the same tip used for interphases characterization therefore the measured values are extremely low (Fig. 3b) and should not be taken into account quantitatively. Fig. 3 shows the evolution of the elastic modulus at the vicinity of a fiber. Measurement was obtained from a single sample, nevertheless, the result presented has been chosen between ten different measurements with negligible dispersion. The AFM tip has a radius of approximately 10 nm, allowing precise quantification along the white dotted line in Fig. 3a. A decrease of around 35% of the modulus can be observed over 200 nm from the fiber/matrix interface (Fig. 3b). This area corresponds to an interphase softer than the matrix. Moreover, average values can be obtained from measurements within a specific area (white dotted squares in Fig. 3a). The Young's modulus used in numerical models is an average value based on ten specific areas at the vicinity of the fiber (e.g. three squares in Fig. 3a).

3. Results

This section is dedicated to validate the model, an RVE with a global mesh size of 1.5 μm is considered in order to obtain accurate results as identified on Fig. 4. All the parameters used for the numerical model are specified in Table 2. A single RVE is used as the fibers distribution does not have significant influence on the composites behavior as discussed in [19]. An extension of this study was made considering ten different distributions; a SD of 351 MPa is identified as well as CV of 3.3%. As this sensitivity is less significant, the choice of a single RVE to predict the macroscopic behavior remains valid.

Table 2. Fixed parameters for mesh sensitivity analysis.

RVE dimensions (μm)	Fibers		Interphase	
	Volume fraction (%)	Diameter (μm)	Thickness (nm)	Elastic modulus (GPa)
150	53	15	200	1.7

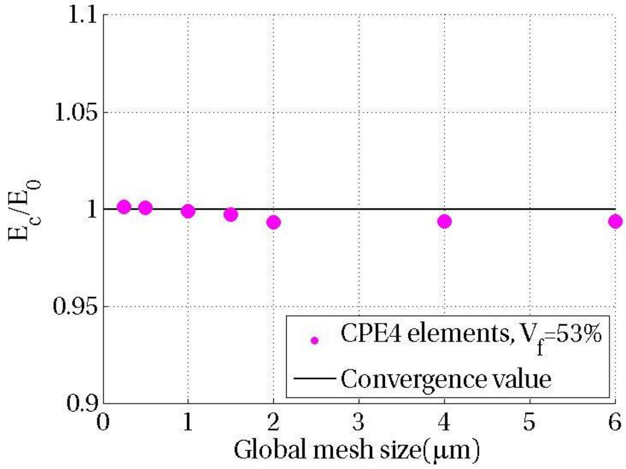


Figure 4. Influence of the mesh size.

For the sake of brevity, the figures of the effects of the interphase thickness, its elastic modulus and its Poisson's ratio on the overall material behavior are not showed here. However, briefs descriptions of these studies are provided. Three different thickness e=200 nm, 300 nm and 500 nm are considered. This range have been chosen based on the experimental measurements and previous work on similar composites. The effective elastic modulus of the composite is evaluated by keeping the RVE dimensions, the diameter of the fibers and the elastic modulus of the interphase as constants with the values showed in Table 2. The composite Young's modulus is inversely proportional to the interphase thickness, which means the composite performs rigidly as the interphase is thinner. This behavior is certainly expected in the case of soft interphases as the matrix volume fraction is been reduced by the interphase volume fraction resulting in an affectation of the global material. Indeed, this effect is a result of the substitution rate of the matrix by this soft interphase. It is also observed that the effect of the interphase thickness becomes more significant as the fiber volume fraction increases.

The effective elastic modulus of the composite was evaluated by keeping the RVE dimensions, the diameter of the fibers and the interphase thickness as constants with the corresponding values showed in Table 2. By decreasing the interphase elastic modulus from 2.7 GPa to 0.7 GPa by increments of 0.5 GPa. The transverse effective Young's modulus increases with the increase of the interphase Young's modulus. A direct proportion is thus identified which means an augmentation of the interphases rigidness is automatically reflected on the overall composite behavior. However, this tendency becomes negligible when the fiber volume fraction is less significant.

Four different Poisson's ratios νi =0.2, 0.3, 0.4 and 0.49 are considered. The effective elastic modulus of the composite is evaluated by keeping the values of all parameters showed in Table 2. The interphase Poisson's ratio has no significant influence on the composite Young's modulus. Indeed, this effective property remains constant no matter which value of interphase Poisson's ratio is considered.

3.1. Experimental/numerical comparison

The stress–strain curve showed in Fig. 8 represent the average mechanical behavior of the composite concerning two numerical models (with and without interphases) and one experimental result (i.e. average value of five repeatable tests). An amelioration of the response is remarked and as expected, the interphase has a positive influence on the prediction of the mechanical behavior of the composite giving a more accurate approach. The result considering the interphase, improved the predictive numerical behavior in 6.8% compared to the result without interphase, this value represents the difference of the elastic modulus of both models (refer to formula 1). As the numerical approach is in the framework of an elastic behavior, a completely linear response is obtained. The absence of damage or fracture phenomenon during the simulation is the reason why the numerical result with interphase does not completely fit the experimental curve. This disparity could be also attributed to the type of elements used for simulation: solid homogeneous.

$$\%Error = \frac{E_{model\_without\_interphase} - E_{model\_with\_interphase}}{E_{model\_with\_interphase}} x\ 100 \quad (1)$$

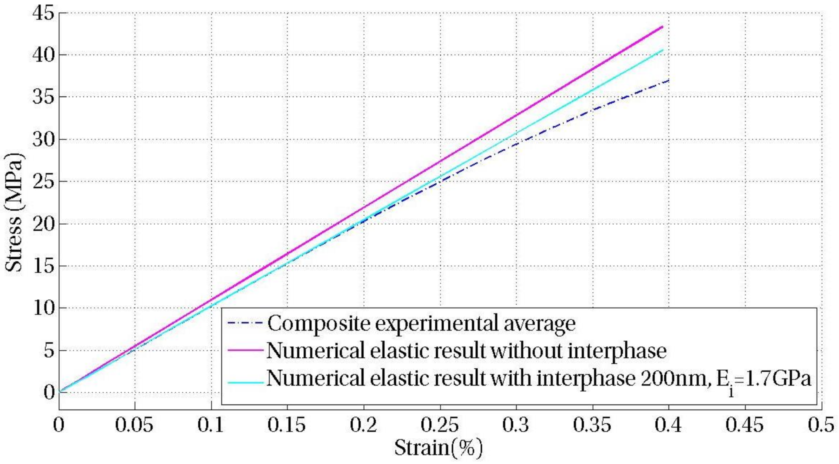


Figure 8. Experimental and numerical comparison.

5. Conclusions

The mechanical behavior of an epoxy matrix composite reinforced with 54% of E-glass fibers is accurately predicted using a FEA homogenization technique on a RVE with random distributed fibers including interphases. Realistic characteristics were attributed to the model: interphases layers of 200 nm with a Young's modulus of 1.7GPa, previously determined experimentally thanks to AFM measurements. Three parameters of the interphase were studied: thickness, Young's modulus and Poisson's ratio. In general, the material properties of a composite along transverse direction are influenced by the interphase properties and this effect becomes more significant as the fiber volume fractions increases. For the present case, a soft interphase is identified experimentally and thus considered in the numerical model, meaning that the Young's modulus of the matrix is greater than the Young's modulus of the interphase. In this context, the composite elastic modulus increases as the interphase becomes thinner. Likewise, an increment on the interphase Young's modulus represents an increase on the composite elastic modulus. To synthesize, a thicker interphase needs higher attention on its elastic modulus as the matrix volume fraction is been substituted so the influence on the global material's behavior increases. Regarding the Poisson's ratio, no significant influence is identified. The experimental/numerical comparison shows an improvement of 6.8% in the prediction of the mechanical behavior of the composite when the interphase is taken into account. Even in the simple framework of linear elastic behavior and even considering solid homogeneous elements, the interphase region plays a significant role when predicting the overall composite behavior. Therefore, further micromechanical approaches should include interphases to better predict the real composite behavior especially when the composite have a high fiber volume fraction. The phenomenon of damage in the composite is apprehended according to the global and local stress fields whether there is an interphase or not.

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