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Integrating fibers and injection molding process variability in short-natural-fiber-reinforced thermoplastics behavior: A review

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Abstract

Injection-molded short-fiber-reinforced thermoplastics are widely used in today's industry. Nevertheless, their mechanical behavior is difficult to model, especially because of strong anisotropy induced by complex fiber distributions of orientation. Moreover, the intrinsic variability of plant fibers' properties leads to an even more complex behavior than with mineral fibers and therefore increase the uncertainty for behavior prediction of these materials. The aim of this review is to provide basic and more specific knowledge about dealing with the uncertainty related to injection molded short-plant-fiber-reinforced thermoplastics behavior, focusing on variability induced by both injection-molded process and natural variability of plant fibers properties. To achieve this goal, it is important to understand the behavior of SFRT before considering the uncertainty induced by the use of natural fibers. Thus, in the first place, the authors have chosen to limit the sources of uncertainty related to fibers by studying the case of a short-glass-fibers-reinforced thermoplastic. Then, after discussing the sources of uncertainty related to the use of natural fibers, the methods for the quantification, the propagation and the management of uncertainties are analyzed.

Keywords: A. Fibers, A. Polymer-matrix composites (PMCs), B. Mechanical properties, C. Statistical properties/methods, D. Microstructural analysis, E. Injection moulding

1. Introduction

The injection molding process is a reversible process which allows manufacturing thermoplastic parts with more or less complex geometry. Compared

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to other manufacturing processes, injection molding has some advantages such as low unit price, thanks in particular to the low cost of thermoplastics and short cycle times, good surface aspect etc. This low-price process can be used to mass-produce small parts (e.g. water bottle top) and bigger parts (e.g. car dashboard). In many cases, this manufacturing process is used for the injection of mineral-fibers-reinforced thermoplastics (e.g. glass fibers, carbon fibers...). More recently, **environmental concerns** oblige industrials to switch to more eco-friendly manufacturing processes [1]. Also, since **the mid of 1970s** [2], **researchers** are interested in the mechanical properties of **natural** fibers aiming at replacing mineral **fibers** by **natural** fibers in injection-molded parts. However, moving from controlled geometries and properties (mineral fibers) to variable ones (natural fibers) implies a less well-controlled mechanical behavior i.e. the variability makes the mechanical behavior more complex to predict.

Therefore, the aim of this review is to do a state of the art of the behavior of short-(glass / **plant**) fiber-reinforced thermoplastics. To do so, it is important for authors to briefly introduce the notion of Short-Fiber-Reinforced Thermoplastics(**SFRT**). Therefore, in section 2, basic **knowledge** regarding the injection molded process (sub-section 2.1), the microstructure of short-fiber-reinforced thermoplastics (sub-section 2.2) and the specificities of **plant** fibers (sub-section 2.3) are **developed**. In order to model short-natural-fibers-reinforced thermoplastics' behavior, it is primordial to understand the complex behavior of **SFRT**. **Thus, in section 3, the thermo-mechanical behavior of thermoplastics reinforced with short glass fibers is studied. In this section, the focus is made on glass fibers due to their controlled geometry and mechanical properties. This allows, in the first place, to neglect variabilities induced by fibers. Thus, the aim of sub-section 3.1 is to present the complex behavior of thermoplastics polymers. Then, fibers' orientation and behavior inside thermoplastics are discussed in sub-section 3.2 and finally the impact of both matrix and fibers on short-fiber-reinforced thermoplastics' behavior is presented in sub-section 3.3.** Then, section 4 is related to the inclusion of sources of variability for the prediction of the mechanical behavior of reinforced materials. The uncertainty quantification is depicted in sub-section 4.1, the uncertainty modelling in sub-section 4.2 and the uncertainty propagation and management in sub-section 4.3. Section 5 aims to discuss the most appropriate tools to handle the uncertainty encountered **with SFRT** according to section 4.

2. Investigation of composites' microstructure

2.1. Basic concept of injection molding

The injection molding machine is composed of a fixed clamping part and a mobile injection part (Fig. 1). The injection process is now a well-known process and is described in [3] :

1) The material granules (pellets) are dosed according to the volume of the mold cavity to be filled and placed in a hopper above the injection machine. In

the case of fiber-reinforced materials, the granules are composed of polymeric matrix and fibers. The granules are heated up to polymer melt temperature and homogenized all along their progress over the rotating screw. The mold, which is closed before the injection, is also heated up to a given temperature;

2) Once the material reaches the end of the screw, the injection can start. The material is injected into the mold through a gate by the transverse movement of the screw. During the filling, the forced flow implies a complex organization for particles in the material;

3) When the mold is filled, the polymer continues to be injected to increase the pressure inside the mold in order to reach a threshold. A high pressure is then maintained on both faces of the mold: this is the packing stage. The aim of this stage is to avoid warpage, by homogenizing polymer shrinkage in the part, and other defects;

4) The final stage consists of cooling the part and ejecting it.

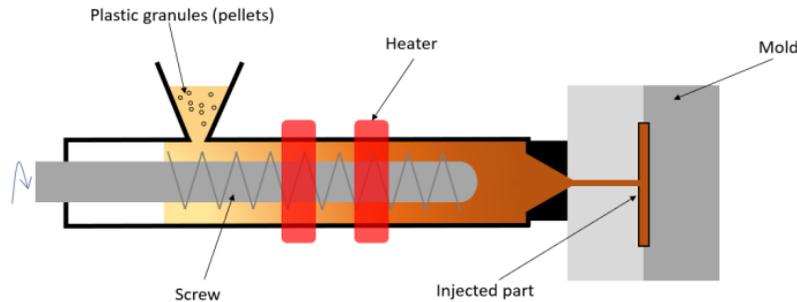


Figure 1: Injection machine

The next paragraph highlights the microstructural organization of SFRT induced by particular phenomena during the injection **molding** process.

2.2. *Microstructure of short-fiber-reinforced thermoplastics after injection molding*

After injection molding, **SFRT** show very complex microstructure, in particular because of the high discrepancy of fiber orientation arising from the forced viscous flow. This discrepancy is governed by material properties (e.g. fiber fraction and geometry), process parameters (injection speed, injection temperature) and part shape.

Yet, mechanical properties of the injected part highly depend on the organization of the microstructure. It is therefore essential to understand the link between parameters of injection process and the microstructure, for given material and part shape, to be able to optimize the process **in order** to reach targeted mechanical properties. Actually, injection molding process parameters highly impact the organization of fibers in composite microstructure because fibers are reoriented following a velocity vector induced by the injected flow. Two types

of flow have a significant impact on the orientation of the fibers [4]. When the material is injected within divergent or convergent geometry into the part, a stretching flow happens and the fibers tend to be perpendicular to the flow. Then, as the material is going through the part, a shear flow happens. This flow allows fibers to reorient themselves parallelly to the injection flow direction. The effect of the shear flow decreases with the distance between the fibers and the walls. It means that fibers at the center of the part (core layer) are less affected by the shear flow and stay therefore mainly oriented transversaly to the injection flow direction.

In addition to those two flows, fountain effect must also be considered. This effect was first noticed with mercury in a glass tube [5] and is characterized by a shear flow induced by the movement of the flow front between solid walls [6, 7]. The viscous flow is then projected from the center of a canal to the edges with a significant impact on the orientation of the fibers [8, 9]. Finally the cold temperature of the mold, compared to that of the flow, together with fountain effect are responsible for the creation of a thin layer near the mold walls where fibers are randomly oriented.

These mechanisms lead to a particularity of SFRT: the “skin-shell-core” structure (Fig. 2), which traduces a strong heterogeneity of fiber orientation through a part thickness. A lot of researchers have described this structure [10, 11, 12, 13, 14, 15, 16]. The main difference is the number of layers that are considered. For Kenig et al. [13] there are nine layers placed symmetrically, while for Bay and Tucker [14, 15] the number of layers is seven. Yet, usually, for injection of thin parts between parallel walls, the most encountered description is a five- (or three-) layers structure, also called “skin-shell (shear)-core” structure [11]. In the case of a three-layers structure, the skin layer is not considered because of its negligible size relatively to part thickness.

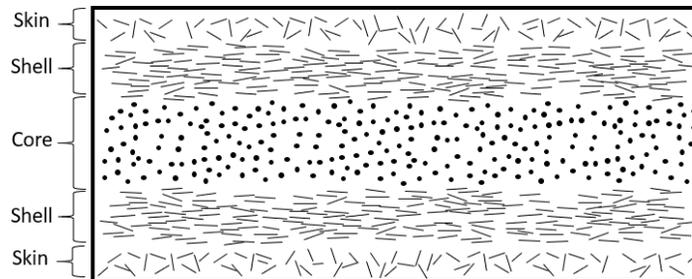


Figure 2: Skin shell core structure

As mentioned before, the fountain effect has a huge impact on fiber orientation. In the skin-shell-core structure, the previous effect is visible in the skin layer, the projection of melted blend against the cold walls prevents fiber to be re-oriented by the shear effect. Then, due to relatively low temperature at mold walls, the fibers are quickly frozen within the polymer and adopt more or

less random orientation. The shell (or shear) layer is composed of fibers mainly oriented parallel to the flow velocity vector, due to the shear effect. Finally, a nearly null shear effect in the middle of the part thickness and the previous stretching flow will make the fibers oriented **perpendicular** to the velocity vector, this is the core layer (see section 3.2).

This kind of microstructure **heterogeneity**, directly stemming from **the injection molding process**, are common to all SFRT, whatever the nature of the **fibers**. Yet, when dealing with **plant fibers**, other kinds of **heterogeneity** directly come from the **intrinsic** variability of **plant fibers** properties. To understand that, it is important to see **plant fibers** as complex structures, to be analysed at several scales, as described in the next section.

2.3. *Plant fibers' composition*

Compared to mineral fibers, natural fibers have relatively low cost, low density, also, they are issued from renewable sources and are biodegradable. On the other hand, while mineral fibers generally have simple and repeatable properties, **natural** fibers can be considered as composite materials with variable characteristics because of their complex structure (Fig. 3). This structure is **currently** divided into different scales with variable morphological parameters. Each scale is also divided into different elements (or layers). At the stem scale (macroscopic scale), the layers are the bark and fibers' bundle. The thickness of the bark and the number of fibers' bundle are variable and potentially different from one stem to another. Then, fibers' bundles **are considered at the mesoscopic** scale and they are composed of a variable number of parallel elementary fibers still glued together which induces variable fiber bundles' section along the bundle. Also, the fact that fibers are glued together introduces fibers decohesion and lead to an early breakage of the fiber bundle. The elementary fibers are **considered at the microscopic** scale and they are single long plant cell. Again, this scale is divided into different cell wall layers. From one elementary fiber to another, the thickness and the diameter of each cell walls are different, leading to even more variable fibers' specificities such as fiber section. Finally, the cell walls are made up with nanoscopic lamellae. Those lamellae mainly consist of cellulose fibrils embedded in hemicellulose and pectin. Fibrils are responsible for the relatively good mechanical strength of **plant** fibers [17]. Therefore, their number and their orientation angle are both important nanoscopic parameters.

3. Thermo-mechanical behavior of short-glass-fiber-reinforced thermoplastics

3.1. *Behavior of thermoplastic polymers*

The macroscopic behavior of thermoplastics results from the behavior of the amorphous phase and, if is present, of the crystalline phase. As a consequence, the crystallinity ratio governs the behavior of thermoplastic. The behavior of

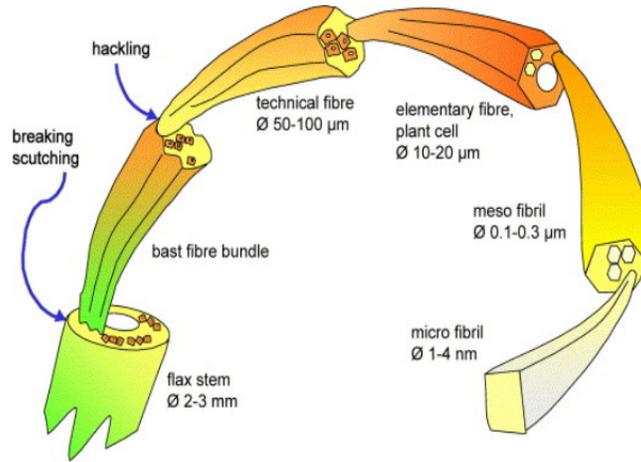


Figure 3: Microstructure of a flax fiber from [18]

thermoplastic polymers highly depends on temperature [19]. Above melt temperature, thermoplastics behave as viscous liquids. Upon cooling, different transitions are characterized by specific temperatures. The crystalline phase thus appears below crystallization temperature, when chains begins to organize themselves. Dealing with the amorphous phase, the glass transition temperature, T_g , delimits the rubbery domain (at high temperature) and the glassy domain (below T_g). The rubbery domain is characterized by a low Young's modulus (E) and a high elastic strain whereas in the glassy domain the amorphous phase is rigid and more brittle. Other important aspects of thermoplastics' behavior are the strain-rate dependency and the temperature dependency within rubbery or glassy domain. Zhang et al. [20, 21] have studied the first aspect for different thermoplastics materials. For instance, the authors have studied the rate dependency of polyethylene (PE) by conducting several tensile tests at constant strain rate [21]. At high strain rate, the PE has a higher Young's modulus than at low strain rate and, more generally, stress level increases with strain rate. For the second aspect, Serban et al. [19] have shown the temperature dependency of polyamide. With Fig. 4, it can be seen that higher temperature reduce the tensile strength of polyamide drastically (from 53MPa for -25°C to 18MPa for 50°C).

In the early strain stages, thermoplastics show a viscoelastic (VE) behavior, i.e. elastic behavior is time and strain-rate sensitive. Then, during a tensile test, the Young's modulus of a VE thermoplastic increases with the strain rate. If the plastic threshold is not reached, the tested specimen retrieves its initial form after more or less long time. It indicates that reversible strain does not vanish instantaneously, contrary to case of elastic behavior, but after a given duration, specific of each thermoplastic. The VE behavior can be represented using rheological models constituted of spring (elastic) and damper (viscous)

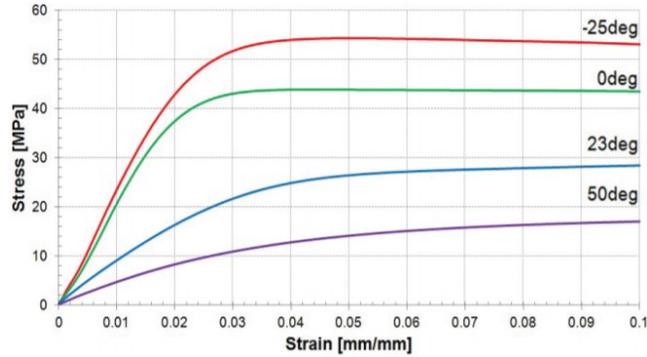


Figure 4: Temperature dependency for polyamide based semi-crystalline thermoplastic ©2016 Serban DA. Published in [19] under CC BY 3.0 license. Available from: <http://dx.doi.org/10.5772/64563>.

elements. Several VE models exist like Maxwell model, Kelvin-Voigt model, Burgers model, etc [22]. The Maxwell model uses a viscous damper and an elastic spring connected in series while the generalized Maxwell model puts in parallel n Maxwell elements and an elastic spring (see Fig. 5).

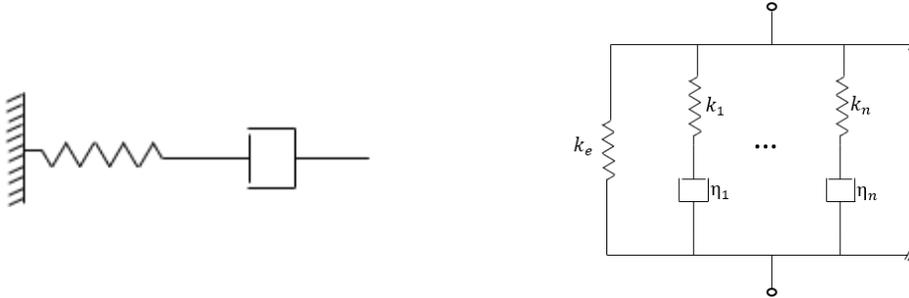


Figure 5: a) Maxwell model b) Generalized Maxwell model

In this model, k_i represent the moduli and η_i the viscosities of each Maxwell element. Each viscous damper has its own characteristic time for strain dissipation. Therefore, the number of Maxwell elements will influence the kinetics of recovery of viscoelastic strain. Accuracy of VE model can be assessed in particular for creep and relaxation tests, with a time-dependent response. During creep tests the material is exposed to a constant stress and the strain response is measured over time. The Fig. 6 shows schematically the differences when using Maxwell model (red line) or Generalized Maxwell model with a finite number of Maxwell elements (black line). As expected, a unique Maxwell element cannot give an accurate prediction of viscoelastic strain/recovery and therefore of creep behavior.

The behavior of thermoplastic materials is also pressure sensitive [23, 24, 25].

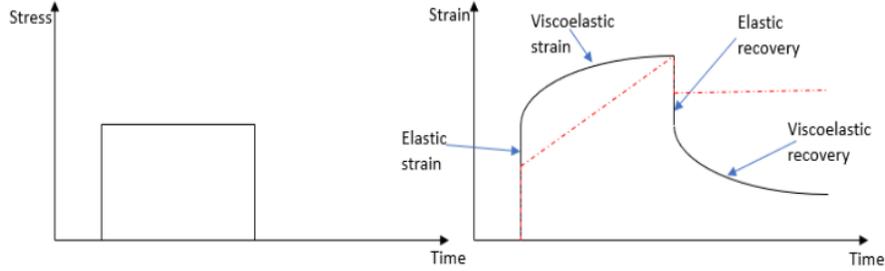


Figure 6: Schematic representation of a creep test and the response of a thermoplastic material; the red dashed-line is the response with Maxwell model and the black line is the response with Generalized Maxwell model

Indeed, those materials show different responses in tension, compression or shear. As a consequence, the isotropic von Mises criterion is irrelevant to describe the yield surface of thermoplastics since it does not consider the effect of hydrostatic pressure. At the contrary, the Raghava yield surface [26, 27] takes the influence of hydrostatic pressure into account by involving the ratio between the initial yield stress in compression and in tension in its expression.

Beyond the viscoelastic domain, thermoplastics plastic flow is also rate-dependent, i.e. behavior is viscoplastic. In addition, several studies [28, 29, 30, 31] have shown the non-isochoric evolution of the plastic flow. For instance, with video-controlled tests, G'sell et al. have observed the volumetric strain for polyethylene terephthalate [29] and quantified the volumetric strain for polypropylene [28]. Finally, thermoplastics generally show a strongly non-linear hardening behavior.

3.2. Short fibers behavior and measurement of their orientation inside the composite

For reinforced polymers several types of reinforcement exist (particles, long fibers, short fibers...). In the case of short fibers reinforcement, the fibers can be mineral fibers (e.g. glass fibers, carbon fibers) or **natural** fibers (flax, hemp, miscanthus, ...).

Mineral fibers are characterized by a cylindrical representation with a ratio length over diameter (e.g. aspect ratio, L/D) much greater than one. In general, their length is lower than one mm in injection molded short-fiber-reinforced composites and their behavior is assumed linear elastic and isotropic or sometimes orthotropic. Compared to the thermoplastic matrix, they have a **higher** Young's modulus (1141 ± 57 GPa [32] against 70-95 GPa [33]).

Plant fibers show more complex mechanical behavior. Indeed, because of the

complex organization of **plant** fibers, their behavior is no longer linear elastic, as mineral fibers, but viscoelastic [34, 35, 36]. Moreover, the complex geometry of **plant** fibers is to be considered. For instance, the cross-section area is generally not circular and varies along fiber length. For more details about **plant** fibers characteristics, see section 3.1.

There are several ways to measure the orientation of the fibers based on 2D or 3D analyses. The 2D observations can be done via scanning electron microscopy (SEM) or optical microscopy. Special precautions must be taken with these methods, especially when preparing the specimens. As for metallic materials, it is essential to polish the specimen and use chemical attacks to enhance the contrast between fibers and matrix. By polishing specimens, there is a risk of degrading the initial cross-section through the grubbing-up of fibers. The main problem with this method is the 2D aspect. Indeed, if fibers were oriented perpendicularly to the plan, their representation with 2D analyses would be a circle. Yet, fibers tilt, with respect to observed plan, is actually variable. Thus, their cut in a 2D view is no longer a circle but an ellipse (Fig. 7). For example, Eberhardt et al. [37], have determined elliptical parameters for different fibers and highlighted the existing significant error induced by this method. They have compared the previous method with results from confocal technique. It has been seen that the error measured depends on fiber orientation and can reach up to 10 degrees of difference with respect to the real value.

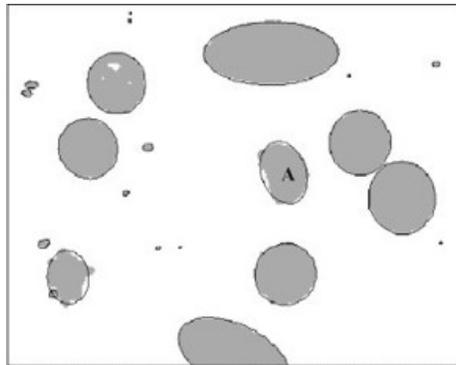


Figure 7: Fiber cross-section identified [37]

When possible, a 3D analysis is therefore preferable. In a growing number of cases, the 3D chosen method is micro-tomography [38]. This method is a non-destructive method, using x-ray scan over a rotating specimen, which allows a slice-by-slice reconstruction of the microstructure volume [39]. As a non-destructive method, it is possible to scan a specimen before testing it, aiming at correlating microstructure organization and mechanical properties. It is also possible to perform in situ micro-tomography tensile test [40] and therefore follow the evolution of microstructure for different loading levels until failure.

3.3. Impact of both matrix and fibers behavior and of their interface on SFRT behavior

Mechanical behavior of short-fiber reinforced thermoplastics depends on the behavior of the two phases (thermoplastic matrix and embedded fibers) but also on the interaction between both phases, i.e. adhesion and behavior of fiber/matrix interface. The complex mechanical behavior of thermoplastic matrix was briefly described in previous section 2.1. Naturally, all its specificities are also encountered in reinforced thermoplastics. For example, Mouhmid et al. [41] have study the strain-rate sensitivity of glass fibers reinforced PA6.6. The authors have shown the same trend as the matrix without reinforcement, i.e. an increase of composite stress level with strain rate (see section 2.1). The case of high strain rate has been studied by Kim et al. [42]. Hopkinson pressure bars were used for the testing of glass fiber reinforced polypropylene. The temperature dependency has also been studied [19, 43]. Thanks to Eftekhari and Fatemi [43] and Nciri et al. [44], one can see the same trend for the glass fiber reinforced thermoplastic composite than for the neat thermoplastic. Indeed, for both cases, the materials become softer with the increase of temperature and more brittle when the temperature decreases. Also, the impact of the variability of fibers on the reinforced material mechanical properties has been studied by Notta-Cuvier et al. [45] for the case of short-flax-fiber polypropylene.

As seen in previous section, fibers have their own behavior that naturally impacts the behavior of the reinforced material. Adding fibers allows to transfer the load initially born by the matrix to the fibers by shear transfer. Understanding the interaction between fibers and matrix is, therefore, crucial for modeling the mechanical behavior of SFRT. A lot of models, for instance shear-lag model initially developed by Bowyer and Bader [46], consider the interfacial shear strength to quantify the adhesion and load transmission at fiber/matrix interface and so on to compute the average stress in a fiber.

The interfacial shear strength (IFSS) obviously depends on the nature of both fibers and matrix. For instance, Yan and Cao [47] have analyzed the differences in microstructure between glass fiber reinforced polypropylene (GF/PP) and carbon fiber reinforced polypropylene (CF/PP). In this study, the IFSS values have been calculated and linked to fiber's critical length. It appears that IFSS increases while critical length decreases. This result agrees with the study of Fu and Lauke [48], the composite strength increases with fiber's length. The values of the IFSS can be much more different if the fibers are natural fiber ($\tau \approx 14.8$ MPa for GF/PP [49] and from four to eight MPa for flax/PP composite [50]) or if the matrix is different ($\tau \approx 5.965$ MPa for CF/PP [47], $\tau \approx 43.8$ MPa for CF polyamide 6.6 [51]).

In the case of natural fibers, the hydrophobicity of the thermoplastic matrix and the hydrophilicity of fibers lead to a poor interfacial adhesion. In order to improve the quality of the interface, chemical or physical treatments can be

used. Enciso et al. [52] have studied the impact of plasma treatment on the interface between flax fiber and polyethylene matrix. They demonstrated that the adhesion was improved with plasma treatment, thus improving the mechanical properties of the composite [52] (e.g. increase of tensile strength). Other treatments have been studied, for example the use of coupling agents like graphene oxide and silane [53]. Also, it is possible to use Maleic Anhydride (MA) grafting to enhance the interface between natural fibers and polypropylene matrix [54]. This treatment is different from other chemical treatments because it affects both fiber surface and matrix. Mishra et al. [55] have shown the increase of mechanical properties (Young’s modulus, flexural modulus. . .) of short-plant-fiber (banana fiber, hemp fiber and sisal fiber) reinforced PP treated with MA (about +13% for tensile strength value). Also, it appears that MA treatment reduces the hydrophilicity of natural fibers.

Through this section, the complex behavior of SFRT has been seen. Indeed, to the specificities of thermoplastics are added the specificities of the fibers, and strong variability in case of natural fibers, and the interaction between the matrix and fibers. In addition to these complex behavior, it is important to remind the significant impact of the injection process on the microstructure of SFRT and by extension on SFRT’s behavior. Therefore, the case of thermoplastics reinforced with plant fibers implies a better understanding and a better modelling of the impact of the injection molded process on SFRT behavior. Also, the geometrical particularities of plant fibers seen in section 2.3 must be considered during the simulations. In order to achieve this goal, the first stage is to find the sources of variability.

4. Sources of variability for composite mechanical behavior

In science, studying uncertainty of a system means studying the variation of system’s outputs (e.g. stress, strain, frequencies, . . .) induced by the lack of knowledge of the system input parameters, such as fibers characteristics, Young’s modulus, etc. In order to control these outputs, it is therefore relevant to know how to deal with uncertainty. First, uncertainty is divided into two classes: irreducible and reducible uncertainties. The difference between them is due to the unpredictable nature of the uncertainty. Indeed, for irreducible uncertainties, even with a large number of data, some events remain unpredictable, on the contrary, reducible (or epistemic) uncertainties may be reduced when dealing with a large number of data. Then, three sources of uncertainty have been highlighted in the literature: measurement uncertainty, model uncertainty and physical variability [56]. Measurement uncertainties are induced by the data measurement stage. For example, it has been seen with Eberhardt et al. [37] that using 2D images for quantifying fiber orientation leads to significant error. This error can be reduced by using 3D measurement instead of 2D measurement (see section 2.2). Also, particular attention must be paid to the means of measurement for sources of uncertainty because it may influence significantly the results of the uncertainty quantification. Indeed, Le Moigne

et al. [57] have observed different results for the measurement of fiber's dimensional properties (e.g. length, cross-section) while using two measurement methods. The first method is purely manual, they have measured the length and aspect ratio (length / diameter) of fibers thanks to a numerical caliper. The second method is made by using a software developed by Microvision Instrument where the geometrical characteristics of the fibers are derived from the detection of the edges of the fibers. With the method "by hand" they measured 300 fibers whereas the software managed to measure 2.500 fibers (Fig. 8). The distributions induced by these two methods are different, in the case of "by hand" measurements, small fibers ($< 60 \mu\text{m}$) are neglected because of eye selection. On the contrary, the software is able to detect all fibers (even the smallest) and give more accurate distribution. Then, the model uncertainty is directly linked with the accuracy of the method. This uncertainty can be reduced by finding the best method to represent the studied case. For example, in their study, Cui et al. [58] have compared three different models and used the maximum likelihood estimation to identify the one in which the model uncertainty is the lowest. Finally, physical variability refer to uncertainty induced by physical variables (e.g. material properties...), environmental conditions (e.g. temperature, humidity...), operational variations (e.g. loading conditions...) or manufacturing process (e.g. injection-molded process...).

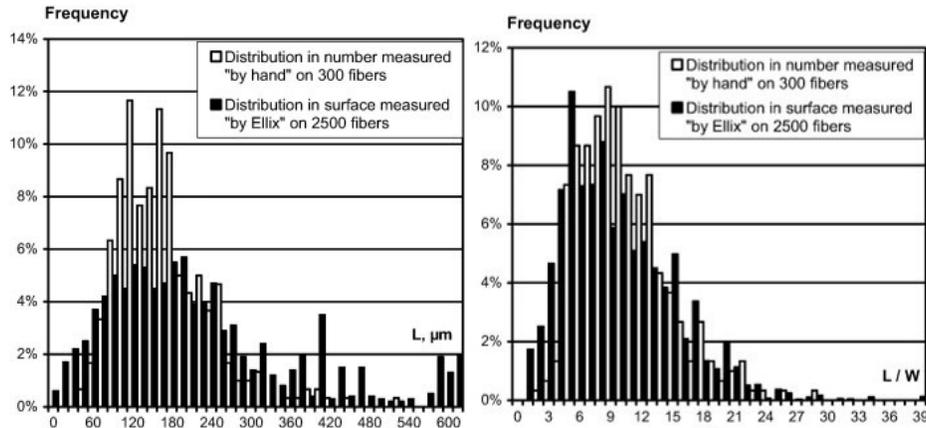


Figure 8: Comparison between "by hand" measurement and measurement made with a software for (a) fibers' length and (b) fibers' aspect ratio [57]

The general procedure to deal with every source of uncertainty requires a first stage of uncertainty quantification and uncertainty modelization, these points are discussed in the next section.

4.1. Uncertainty Quantification for composite materials from micro-scale to macro-scale

Composite materials are multi-scale material thus, the uncertainty may arise at any scale, to the micro-scale (material properties), as well as the meso-scale (fiber orientation) and macro-scale (geometry, composite properties).

Micro-scale quantification for composite materials

In SFRT, the matrix is usually the component with the highest volume proportion (55wt.% to 70wt.%). However, the uncertainty of its mechanical property **does not have** a significant impact on composite uncertainty due to much lower stress levels. On the contrary, with a lower volume proportion and depending on loading direction, fibers have more significant impact on composite uncertainty. Synthetic fibers (e.g. glass fibers) have constant mechanical properties thanks to a controlled manufacturing process. On the contrary, **plant** fibers present much more variable characteristics leading to bigger uncertainty for composite's behavior. This uncertainty is induced by several aspects such as the growing conditions of the stem, the morphology of the fiber or even chemical composition of fibers. Thuault [59] highlighted the importance of the cultivation conditions during the growing of the plant by comparing mechanical properties of flax cultivated in 2005 and in 2008. For the first one, climatic analyses have shown optimal temperature and sufficient water intake. For the second one, the same analyses have found a wet period during harvesting. The test highlighted better mechanical properties for the flax cultivated in 2005 (e.g. $E \approx 55 \pm 25$ GPa for 2005 and $E \approx 40 \pm 15$ GPa for 2008). Moreover, the location of the extraction of fibers inside the stem impacts the mechanical properties of **plant** fibers. Indeed, several studies have measured the variability of fibers' diameter over its length [60, 61, 62]. Charlet [62] has shown that extracting fibers from the middle of the stem lead to different fiber properties. For example, the mean diameter for flax fibers extracted from the middle of the stem is $12,4 \pm 3,2$ μm against $16,5 \pm 5,1$ μm for fibers extracted from the bottom of the stem. The difference is due to better amount of cellulose and pectic cement in the middle of the plant. Baley [63] **has** studied the importance of fiber diameter on the Young's modulus of flax fibers (Fig. 9). The results link a decrease of the Young's modulus when fiber diameter increases. The same trend has been highlighted for fiber length by Anderson et al. [64], the strength increases while fiber length decreases. For authors, this phenomenon is linked to fiber defects content. Indeed, the longer the **fibers** are, the higher the occurrence of defects is.

Also, the link between mechanical properties of the fibers and their composition **has** been pointed out by many studies [18, 57, 63, 65, 66]. Indeed, it has been noted that fiber strength increases as cellulose content increases. Furthermore, Kaack et al. [66], managed to link the Young's modulus of miscanthus fibers with its chemical composition in terms of lignin and cellulose. For the morphology of fibers, the number of elementary fibers within a fiber bundle is also variable. Indeed, Mattrand et al. [67] have studied the number of

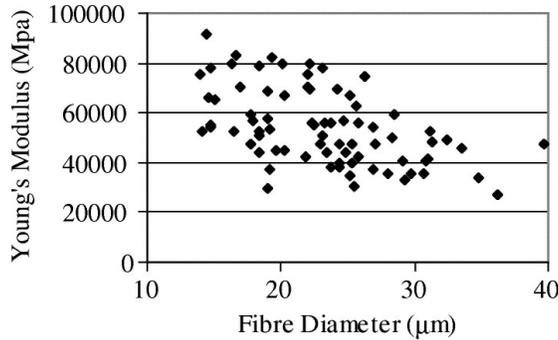


Figure 9: Young's modulus of flax fiber [63]

elementary fibers included in flax technical fibers. They found that the minimum number of elementary fibers was two and the maximum was fifteen. The dimensions of elementary fibers are also to consider. Because of their complex structure and geometry, it is difficult to define proper values for fiber cross-section (or diameter). Thomason et al. [60] have compared values of cross-section diameter directly measured on fiber bundles versus values of cross-section diameter calculated by assuming a circular shape for the cross-section of flax fiber bundles. According to the authors, the cross-section values obtained with this method are the double of values obtained when measuring the cross-section directly on fiber bundles. Therefore, the method assuming a circular fiber bundle cross-section is not an efficient method because it leads to wrong values of fibers' Young's modulus due to previous error.

Meso-scale quantification during injection molded process

The meso-scale of composite materials refers to fiber orientation. The orientation of fibers is set during the injection molded process (see section 1.2) and may be influenced by several process parameters.

The gates directly impact the injection of fibers into the mold, and thus their orientations [68, 69]. The formation of the core layer, with fibers oriented transversely to the Injection Flow Direction (IFD), is made by a diverging flow induced by gating arrangements. However, film-gates or sprue-gates allow fibers in the core layer to be oriented parallelly to the IFD [70].

The injection speed has a direct effect on the thickness of the core layer, and thus on fibers' distribution of orientation through the thickness [71]. Actually, Gupta and Wang [10] have shown that decreasing the injection speed increases the thickness of the skin layers, then leading to a translation of the shear layers to the core, thus finally reducing the thickness of the core layer. On the contrary, increasing the injection speed increases the thickness of the core layer. The studies of Shokri and Bhatnagar [72, 73] show the impact of the packing stage on fiber's orientation. Indeed, they have shown that an insufficient packing stage

duration leads to an increase of the core layer. Nevertheless, few studies have evaluated the impact of the packing stage during the injection molded process. The mold temperature regulates the skin layer. A high gradient between temperature of mold walls and melted flow will quickly freeze the material near the walls. So, fibers do not have the time to re-orient themselves and adopt random orientation. It can be noted that the thickness of those skin layers increases with the gradient of temperature. However, with a low temperature gradient, the material near mold walls is slowly cooled and fibers can be re-oriented by the fountain effect [71, 74]. Also, the melt temperature impacts the shear rate and thus the fiber orientation by means of the rheology of the material. A reduction of this temperature induces an increase of the viscosity of the material and a faster solidification of the melt near the mold walls, thus increasing the shear rate. Therefore, fibers have time to re-orient themselves and the thickness of the shear layer increases, i.e. more fibers are oriented parallel to the IFD [75]. Moreover, the rheology of the material affects the velocity profile during filling [76, 77]. Thus, the fibers follow the same pattern and are re-oriented, i.e. the structural organization of the final product arise from the velocity profile during filling phase.

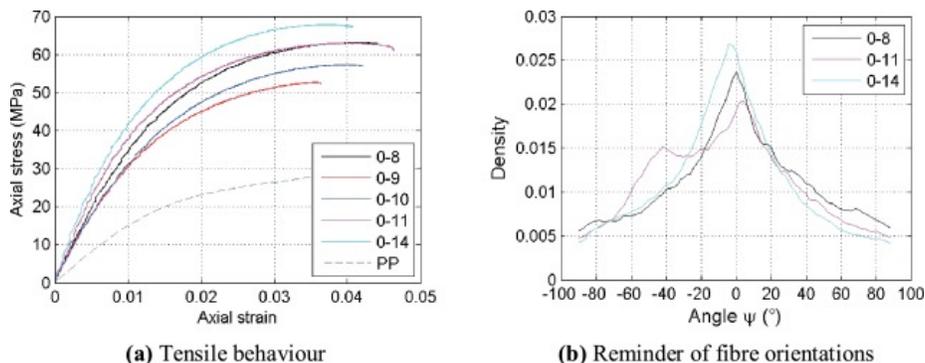


Figure 10: Tensile test of glass fiber reinforced polypropylene [78]

Keeping in mind that **fiber** local orientations have a great impact on SFRT mechanical properties, it is therefore crucial to be able to quantify the variability of fibers orientation to control the mechanical properties of SFRT. Indeed, the wide range of fibers orientation is responsible for an anisotropic behavior of the composite. An accurate prediction of composite behavior therefore requires knowledge of fibers' orientation. Notta-Cuvier et al. [78, 79] have studied the dispersion of fiber orientation inside tensile specimen issued from a same cutting angle, as illustrated by Fig. 10. In this figure, one can see that the dispersion of tensile results (Fig. 10 a) is directly linked with the orientation measured inside the scanned volume (Fig. 10 b).

Macro-scale quantification for composite materials

The studies of Vincent et al. [68] and of Gillespie et al. [80] highlight the dependency of the shear rate to the thickness of the pieces. According to Vincent et al. [68], in the case of thin pieces, fibers' orientation is nearly uniform because of high shear rate. This high value allows fibers to re-orient themselves parallelly to the IFD. On the contrary, when the thickness of pieces increases, the fibers cannot be re-oriented parallelly to the IFD because of low shear rate. The Fig. 11 shows fiber's orientation throughout several thicknesses of injection molded plaques (200mm x 40 mm). As it can be seen, by increasing the thickness of the pieces a core layer emerges and becomes larger with the increasing thickness (i.e. more fibers oriented perpendicularly to the IFD).

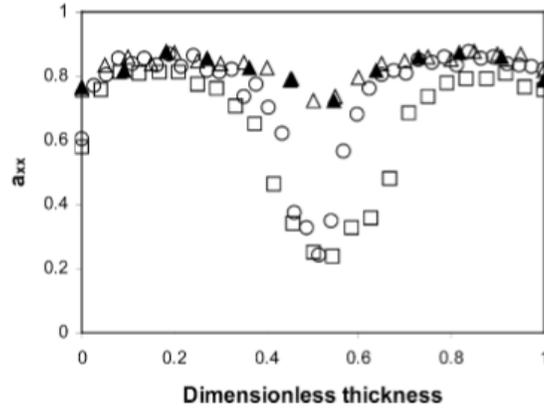


Figure 11: Evolution of the fiber orientation through thickness. The bold triangles represent parts with thickness of 1.1 mm, the empty triangles for 1.7mm, the circles for 3mm and the squares for 5mm [68]

Fiber content in the composite also has an influence on the orientation of the fibers [81] and on the mechanical properties of the composite material [48, 81, 82, 83]. For Bernasconi and Cosmi [83], the higher the fiber content, the higher the number of fiber oriented parallel to the IFD is. In addition to previous parameters, it is worth noting that composite behaviors seen in section 2.3 (such as temperature dependency, strain rate dependency...) must also be considered for the uncertainty quantification in order to include composite behavior dispersion [84].

Table 1 summarize the uncertainty found for composite materials seen in section 4.1.

4.2. Modelling uncertainty of composite materials

After identifying the sources of uncertainty and their relative amount for composite materials, the next stage is to model them. This step consists in

Type	Uncertainty kind	References	Identified Uncertainty
Plant	Cultivation conditions	[59]	Fiber's mechanical properties
	Stem chemical composition	[60, 61, 62]	Fibers' mean diameter, Young's modulus
Process parameters	Fiber geometrical properties	[63, 64]	Young's modulus
	Gates design	[68, 69, 70]	Fiber orientation in core layer
	Injection speed	[10, 71]	Thickness of the core layer
	Packing stage duration	[72, 73]	Thickness of the core layer
	Mold temperature	[71, 74]	Thickness of the skin layer
Melt temperature	[75, 76, 77]	Rheology of the material	
Material characteristics	Rheology of the material	[76, 77]	Material velocity profile during injection
	Fiber content	[48, 81, 82, 83]	Thickness of the core layer
Other	Means of measurement	[60]	Precision of values
	Thickness of the pieces	[68, 80]	Shear rate

Table 1: Summary of the different sources of uncertainty enumerated in sub-section 4.1

converting scattered experimental data into non-deterministic quantities. To achieve this aim, several theories and methods are used. Probabilistic theories, interval theories and fuzzy sets theories are traditionally the most used in mechanical engineering. For composite problematics, authors have mainly investigated the probabilistic way by defining a probability density function from experimental data. Two probabilistic methods can be found in the literature, namely parametric and non-parametric. In the case of parametric probabilistic approaches, the aim is to quantify the uncertainty of the variable parameters such as material properties, for example fibers' Young's modulus. To do so, random variables are associated to each uncertain parameters. For example, Hohe et al. [85] used this way for long fiber reinforced thermoplastic and **showed** the variation of the material response induced by the uncertainty of fiber orientation and local fiber content. In the case of non-parametric probabilistic approaches, the matrices governing the problem are considered uncertain to de-

fine the uncertainty of the whole system. In their study, Piovan et al. [86] have used both parametric and non-parametric approaches for a dynamic study with thin-walled beams defined with a graphite-epoxy composite (AS4/3501-6). For the parametric probabilistic approach, the authors have considered the parameters related to the elastic properties as variable whereas uncertain stiffness and damping matrices of the model are defined for the non-parametric approach. One conclusion of their study is the presence of areas that are more sensitive to the variation of parameters (parametric approach) and/or to the variation of the model (non-parametric approach). Hessman et al. [87] developed a toolbox in order to extract probability density function for fibers' properties (e.g. Gaussian distribution for fiber radius). Also, Moigne et al. [57] tried to find the most suitable probability density function between Weibull distribution and log-normal distribution for flax and sisal fiber length. Both distributions were evaluated with P-value analysis. The aim of this evaluation is to characterize the likeness between experimental data and the chosen distribution. According to the authors, Weibull distribution shows best fitted results in that case.

In the case of sparse data, the definition of probability distribution is questionable. Indeed, in numerous articles, authors have arbitrarily chosen uniform laws to model the input uncertainty. In this context, non-probabilistic approaches, for example fuzzy or interval theories [88, 89] are considered. In this case, the variability is defined by an interval or a set of intervals. For the fuzzy set theory, the reference method is the Zadeh's Extension Principle (ZEP). The aim is to build the membership functions of output data as a function to those of input data. Fuzzy set theory is an extension of ordinary set theory, in which each object either belongs to a set or does not. Fuzzy set theory introduces the notion of degree of membership, using a membership function (triangular, trapezoidal. . .) to describe, for each element in the domain, the level of membership in the fuzzy set. This function can take different forms, depending on the user's perception of the input's imprecision (Fig. 12). Dey et al. [88] used the fuzzy theory for the representation of ply orientation angle, graphite-epoxy elastic modulus and shear modulus. Naskar et al. [90] have worked on a fuzzy representative volume element in order to add the spatial variation of parameters such as ply orientation. Babuska et al. [91] have developed a hybrid fuzzy method which combines the traditional fuzzy moments with random fields. This new method aims to predict the uncertainty of fiber composites (more broadly in multiscale models) more accurately than traditional fuzzy logic. From these studies, it can be seen that the general representation, or at least the most used, for the fuzzy membership function is a triangular function. This function usually associates the mean value with the apex of the triangle and the extremities of it with the standard deviation of the measured data (see Fig. 12). Therefore, it is legitimate to ask if this representation is the most relevant. Indeed, as the measured data are part of the samples, their value from the membership function could be equal to one, instead of a decreasing value from one to zero. Then, the uncertainty added by the measuring stage, the model used, etc, should be taken into account by adding unmeasured values. This inclusion

is made with the membership function, by decreasing the values of this function with a user-defined slope. A trapezoidal function is created by doing this method [92]. The fuzzy logic used by Sodoke et al. [93], aims to accommodate the Young's modulus measured with different test rig (tensile test, bending test and acoustic impulse). During the test rigs, the material used was a flax-epoxy natural fiber composite.

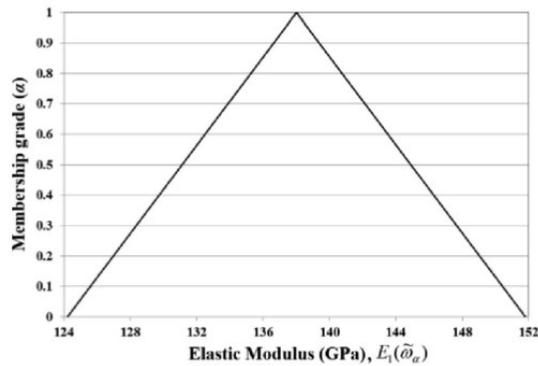


Figure 12: Example of fuzzy membership function [88]

Some authors have studied the results given by different types of uncertainty-based methods. Alazwari and Rao [94] have studied model uncertainty for stress distribution through thickness of laminates composites by using probabilistic approach, interval analysis and universal grey theory in order to compare the results. In universal grey system theory the arithmetic operations are different from interval theory. The aim of this method is to avoid the dependency problem encountered by the interval theory which leads to more accurate results [95]. It can be seen that in this case, the confidence interval given by the probabilistic approach is more accurate than the one given by interval analysis and universal grey theory. This accuracy is provided by a better precision of the probability distributions used in the probabilistic approach.

Table 2 summarize the different methods to model the uncertainty for composite materials seen in section 4.2

After modeling the uncertainties, the next stage is to propagate the uncertainty of the input(s) to the output(s). This last stage is depicted in the following section.

4.3. Uncertainty propagation and uncertainty management

Like in other domains, the aim of uncertainty propagation step in mechanics, is to transfer the uncertainty associated to input variables (material, geometry, ...) towards the output ones (displacement, strain, stress, failure, frequency, ...). The link between the inputs and outputs is made by a transfer function,

Model	Type of approach	References	Comments
Probabilistic	Parametric	[86, 85]	Graphite epoxy composite, Short-glass-fiber-reinforced polyamide
	Non-parametric	[86]	Graphite epoxy composite
	PDF extraction	[87, 57, 94]	Short-glass-fiber-reinforced thermoplastic, Natural-fiber-reinforced composite
Fuzzy Logic	General Fuzzy Logic	[88, 93]	Triangular function, Young's modulus measurement
Fuzzy	Fuzzy representative volume element	[90]	Aim to add spatial variation
	Fuzzy-stochastic	[91]	Combination of fuzzy logic and random fields
Interval	Interval theory	[94, 95]	Comparison of different methods
Other	Universal Grey Theory	[94]	Comparison of different methods

Table 2: Summary of the different way to model the uncertainty enumerated in sub-section 4.2

which directly traduces the studied case and so the mathematical problem to be solved. This "transfert function" can be analytical, numeric or a succession of linear or nonlinear stages, integrating for example finite element computation models.

For the uncertainty propagation, Monte-Carlo Simulation (MCS) is the reference probabilistic method. The sampling MCS is widely used with finite element method [96, 97, 98] . The principle is to evaluate the "transfer function" for a large number of samples of the studied model while applying a FEM simulation. The used values for input variables are randomly extracted through associated Cumulative Density Functions (CDF). For example, Lee et al. [99] and Zhou et al. [100] have respectively estimated mechanical properties of glass fiber reinforced composite and simulated tensile failure process of carbon reinforced

composite. Next, Jeong et al. [101] used MCS to investigate tensile failure process of fiber reinforced plastic laminated plates. However, to achieve a convergence for the mean or the variance of outputs and guarantee a good level of calculation, the number of samples must necessarily be very important and so the method becomes time-consuming.

To overcome the computing time problem, alternative approaches have been proposed in the literature. On one side, Sudret [102, 103] distinguishes two probabilistic models, intrusive and non-intrusive methods. The first one requires a modification of the computing code and the definition of a new mathematical problem to identify the unknowns of the proposed algorithms. On the contrary, the second one makes reference to a way where the initial code is not modified and is used to generate a training set. In the literature, many authors used non-intrusive methods. Indeed, for Carrere et al. [104] non-intrusive methods are the answer of a large number of application, such as industrial problems, in which the computing code cannot be modified. Two methods stand out, the Reduced Order Models (ROM) and the surrogate models, either directly integrated in the alternative method (for example in the case of **Polynomial Chaos Expansion, PCE**, with polynomial series), or coupled with the reference method (for example a kriging with MCS). The idea of the ROM is to reduce the initial database (e.g. proper orthogonal decomposition). Then, the aim of surrogate models is to replace the initial problem by an efficient approximation of output solutions (e.g. PCE, kriging). These last years, PCE methods have been widely used and especially the generalized PCE. The principle of this method is to project stochastic solutions of the initial model onto polynomials orthogonal base [105]. According to the Askey scheme, the type of polynomial series is defined by the type of the variable (for example, for Gaussian variables, Hermite polynomials are used [106]). In their study, Thapa et al. [107] proposed a non-intrusive PCE method, which relies on an automatic differentiation to facilitate the calculation of sensitivities of the stochastic response and the expansion terms with regard to the uncertain input parameters. This proposed method was exploited to investigate multi-scale buckling in the case of carbon fiber reinforced composites.

The most efficient alternative methods are firstly based on the decomposition of the input membership functions into intervals according to α -cut level. Secondly, each interval problem is transformed into a min-max optimization problem and a surrogate [108], or a ROM [109], is integrated at each iteration. This strategy has been successfully used by Massa et al. for different mechanical problems (static [110], modal [108, 92], frequency responses [108], stability analysis [109]). Dey et al. [88] used a new fuzzy propagation method based on the work of Chowdurry et al. [111], for dynamic characterization of a laminated composite (first three natural frequencies).

In order to better understand the behavior of model output(s) throughout variation of input parameter(s), sensitivity analyses are classically used. These analyses quantify the weight of each input parameters by using results from Uncertainty Quantification (UQ) and Uncertainty Propagation (UP) stages.

Global sensitivity analysis allows to simulate simultaneous variation for each parameter during the analysis. On the contrary, during local sensitivity analysis the variation of parameters is made one by one and does not include interactions between parameters. In order to find the key parameters (i.e. parameters having the greatest impact), it is essential to compare simultaneously multiple input parameters and therefore to use global sensitivity analysis. Del Mastro et al. [112] have conducted a numerical analysis study in order to find parameters with the biggest impact for flax and hemp fibers mechanical properties. At the beginning of the study, twenty-six different parameters were selected (such as fiber diameter, viscoelastic parameters, elastic moduli of fibers' constituents...) and then, with two sensitivity analyses, they managed to reduce to seven key parameters. For the first sensitivity analysis, they used Morris screening method. This method evaluates the impact of one input parameters at a time on the output. This method is usually used for models with numerous input parameters. The aim is to reduce the dimension of the problem with a first selection made by calculating a sensitivity index (i.e. elementary effect). Then, the second sensitivity analysis used is the Fourier Amplitude Sensitivity Test (FAST). The latter is a variance-based method, it means that the variance of input parameter(s) is used to determine the sensitivity indices. Therefore, attention must be paid to the estimation of inputs' variance.

Ghauch et al. [113] combined PCE with Sobol indices to achieve sensitivity analysis with the injection of fiber composite materials. They have studied the impact of 74 parameters on the injection to identify the parameters that are the most critical during the injection process. This study leads to a better understanding of the injection process and makes possible the optimization of injection process. Sobol indices are ANOVA based methods (ANalysis Of VAriance). The aim is to decompose the total variance of the output(s) into indices that reflect the impact of each inputs on the output(s) and also the interaction between inputs. Omairey et al. [114] have made sensitivity analysis in order to analyze the effect of multi-scale uncertainty on reliability of the composite. They used Sobol indices for the sensitivity analyses and linked composite mechanical properties (Young's modulus, shear moduli and Poisson's ratio) to material and geometrical (both fiber and matrix) uncertainties. It has been shown that the Young's modulus E_{11} is mostly affected by fiber's stiffness. This result agrees with the assumption made in section 2.2: fibers efficiency is maximized if they are loaded in their axis direction.

Another way to manage the uncertainty is to use the Robust Design Optimization (RDO). The aim of this method is to find the best compromise between characteristics of the system. The RDO method have been used by Antonio et al. [115] with angle-ply composite made of E-glass/epoxy. In their study, they wanted to optimize the couple performances of the system and its robustness. In addition to the RDO method, they used Pareto curves to determine the best couple. Also, Kalantari et al. [116] used the RDO in order to optimize the strength, the cost and the weight of carbon/GF reinforced hybrid composite during flexural loading. In their study, they have used this method by considering the uncertainty induced by inputs parameters such as thickness, fibers'

Model	Type of approach	References	Comments
Reference method	Monte Carlo simulation	[96, 97, 98, 99, 100, 101, 117]	Reference method
Alternative methods	surrogate Intrusive / non intrusive methods Reduced order model Fuzzy propagation	[108, 105] [102, 103, 104] [109] [88, 111]	PCE or kriging min-max optimization problem and ROM dynamic characterization
Sensitivity analyses	Morris screening Fourier amplitude sensitivity test (FAST) Sobol indices	[112] [112] [113, 114]	flax and hemp fibers mechanical properties flax and hemp fibers mechanical properties ANOVA method
Optimization	Robust design optimization	[115, 116]	find the best compromise between characteristics

Table 3: Summary of the different way to propagate and to manage the uncertainty enumerated in sub-section 4.3

orientation.

Through this section, different sources of uncertainty and different way of reporting this uncertainty have been seen. Also, the propagation of the uncertainty from different elements (such as fibers, process parameters...) to SFRT behavior has been reviewed. Therefore, one can see that integrating natural fibers in SFRT simulation is not an easy task. Indeed, to the microstructure variabilities induced by the injection molded process are added the geometrical variabilities of **natural** fibers.

Table 3 summarize the different **methods used** to propagate and to manage the uncertainty for composite materials as seen in section 4.3

5. Conclusion

The use of **plant** fibers in SFRT has grown over the last years because of environmental concerns and public opinion. Thus, being interested in the study for modeling the behavior of thermoplastics reinforced with short vegetable fibers seems both legitimate and necessary.

In order to model SFRT behavior, it is first important to clearly understand the behavior of the matrix and fibers separated. Indeed, the matrix implies a viscoelastic behavior, a temperature dependency and a strain rate dependency.

On the other hand, adding fibers implies an anisotropic behavior for the SFRT and a better control of the injection process (due to fibers' orientation).

This review has highlighted the significant roles of the uncertainty quantification, the uncertainty modelling, the uncertainty propagation and finally the uncertainty management. Through the section 4, different tools have been seen for the determination of the uncertainty of fiber-reinforced thermoplastics and for integrating different sources of uncertainty into their modelization. It has been seen that few papers concern the use of SFRT materials. Thus, according to the authors, the uncertainty quantification of SFRT materials should be done by using 3D measurement methods because of its non-destructive aspect. For the propagation of uncertainty, non-intrusive methods such as surrogate models (PCE) are the most relevant choice. Finally, the management stage is context-dependent and it is difficult to provide a universal method. Nevertheless, two methods stand out, the sensitivity analysis and the robust optimization. Sobol indices (section 4.3) allow one to find the parameters with the highest impact on material properties. Then, the second method, the robust optimization, is probably more interesting in an industrial context. Indeed, this method can be used to rethink an injected part in order to optimize its cost, its weight, etc. This review has shown that further researches are required for SFRT material. Significant research is currently being carried out by the authors aiming both the determination of uncertainty induced by the fibers and by the injection process and the integration of uncertainty during numerical simulations for thermoplastics reinforced with glass fibers and **natural** fibers.

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