INFLUENCE OF MANUFACTURING PROCESS OVER MECHANICAL PROPERTIES OF DISCONTINUOUS CARBON/EPOXY COMPOSITES REALIZED BY INJECTION-TRANSFER

T. Poumadère[1, 2*], F. Lachaud[2], G. Bernhart[2], R. Piquet[2], F. Berthet[2]

1 : Equip'Aéro Technique, Z.I. Buconis, 32600 L'ISLE JOURDAIN, France
2 : Université de Toulouse ; INSA, UPS, Mines Albi, ISAE; ICA (Institut Clément Ader), 10 Avenue Edouard Belin, 31055 TOULOUSE, France
**E-mail : t.poumadere@equipaero.com, t.poumadere@isae.fr*

Keywords: discontinuous long fibers, injection-transfer process, tensile test, process optimization

Abstract

This study presents the influence of the manufacturing parameters of the injection-transfer process over tensile properties of Discontinuous Carbon Fiber Reinforced Composite (DCFRC) manufactured by this process. The matrix is epoxy and the fiber volume rate is 56%. The material studied is considered as a long fiber composite since fibers are longer than 1mm. Design of experiment method has been used to achieve this aim. Moreover damage of the material has been studied and the main fracture mode of the material could be identified. A numerical model was also developed to predict the elasticity modulus of the material.

1 Introduction

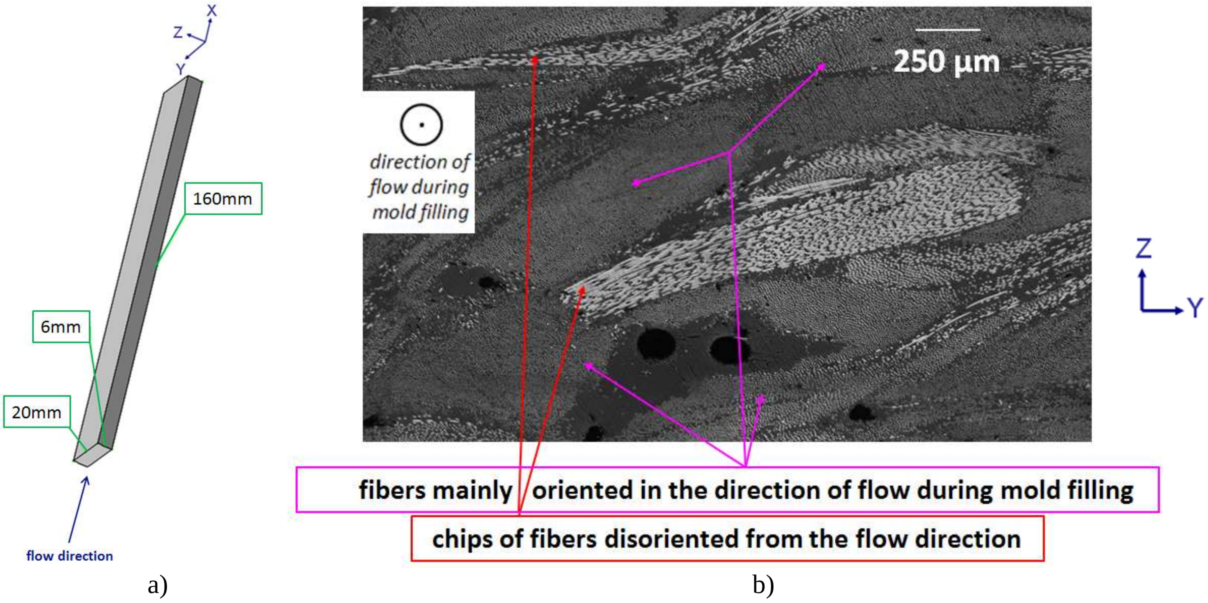
Very few research have been carried out on discontinuous long fibers (longer than 1mm) and thermosetting resin composite material manufactured by injection molding. The French society Equip'Aéro Technique developed a new composite manufacturing process called PIMOC which is an injection-transfer process. It consists in injecting into a closed mold chips of unidirectional carbon fibers/epoxy resin prepreg. Thus, 3D parts made of Discontinuous Carbon Fiber Reinforced Composite (DCFRC) can be manufactured. Fibers are longer than 1mm and oriented in the three directions of the space contrary to typical discontinuous composites manufacturing processes (SMC and HexMC compression molding).

Short fibers composites (smaller than 1mm) with thermoplastic matrix are well known from a manufacturing point of view as well as concerning their mechanical properties [1][2][3]. Nevertheless, there are very few works on discontinuous long fibers composites with a thermosetting matrix manufactured by injection [4].

The aim of this study is to determine the influence of the manufacturing parameters of the injection-transfer process over mechanical properties of DCFRC. Four main parameters have been identified: Injection Speed, Fiber Length, Viscosity of the resin during injection and Holding Pressure applied during crosslinking of the resin. Non Orthogonal Latin Hypercube (NOLH) design of experiment (DOE) has been used to achieve this aim.

2 Materials and manufacturing process

The material used is made of high strength carbon fibers and epoxy resin. The studied samples are rectangular (160mmx20mmx6mm) and made by injection-transfer process (cf Figure 1.a). Their fiber volume rate is 56%. The Figure 1.b shows the typical organization of fibers in rectangular sample.



a) b)

Figure 1. Studied material: a) Rectangular samples studied - b) Microstructure of the DCFRC material manufactured by injection-transfer process (Optical Microscope)

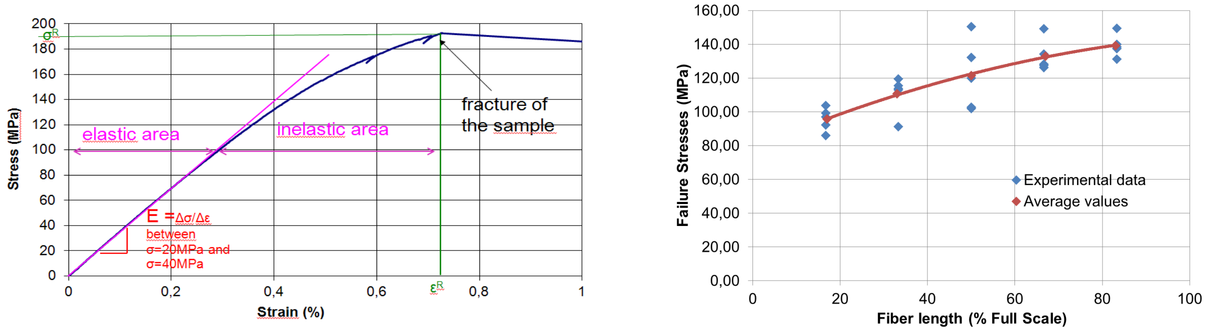
The planigraphic plane is perpendicular to the flow direction during the filling of the mold. It is clear that fibers are organized in clusters or chips of fibers that can have a different orientation from the flow direction (X-direction on Figure 1.a). However, the major part of fibers is oriented in the direction of the flow.

3 Study and results

3.1 Influence of manufacturing parameters of the injection-transfer process over tensile properties

The mechanical tests have been performed on a 100kN tensile/compression Instron testing machine. Glass/epoxy tabs have been previously stuck on samples presented on Figure 1.a. For each configuration, five rectangular samples have been tested. These tensile tests were carried out with an imposed displacement (0,5mm/min until fracture). Strain toward the direction of filling of the mold (X-direction) was recorded thanks to a laser extensometer. That means makes it possible to record a macroscopic strain (l_0 = 80mm).

Figure 2.a presents the typical tensile behavior of the studied material. Failure stress (σ^R) and failure strain (ε^R) have been calculated in each case as explained. The modulus of elasticity (E) of samples have been calculated on the linear area of stress-strain curves, between σ = 20 MPa and σ = 40 MPa. Previous studies investigated the influence of fiber length over the failure stress. The values of manufacturing parameters used to make the samples are confidential. Consequently each parameter is expressed as a percentage of the full scale (F.S.) instead of being explicitly expressed in this paper. Figure 2.b represents the evolution of failure stresses toward fiber length for samples manufactured with the same parameters. The injection speed used corresponds with 8% of the full scale.



a) b)

Figure 2. Influence of fiber length over tensile failure stress of the DCFRC material manufatured by injection-transfer process : **a)** Typical strain-stress curve - b) Results for injection speed corresponding with 8% F.S.

Tensile failure stress obviously increases as the fiber length increases. The average values calculated among the five samples for each case are included between 95 and 150 MPa. NOLH DOE was used to determine the exact influence of each manufacturing parameters (injection speed, fiber length, viscosity of the resin and holding pressure) over mechanical properties. This method decreases the number of experimental configurations compared with factorial design like Taguchi's tables. That DOE included 17 configurations and all parameters change at each configuration. Five samples were manufactured for each configuration and tensile tests were performed. Figure 3 represents the variations of failure stress toward injection speed and fiber length.

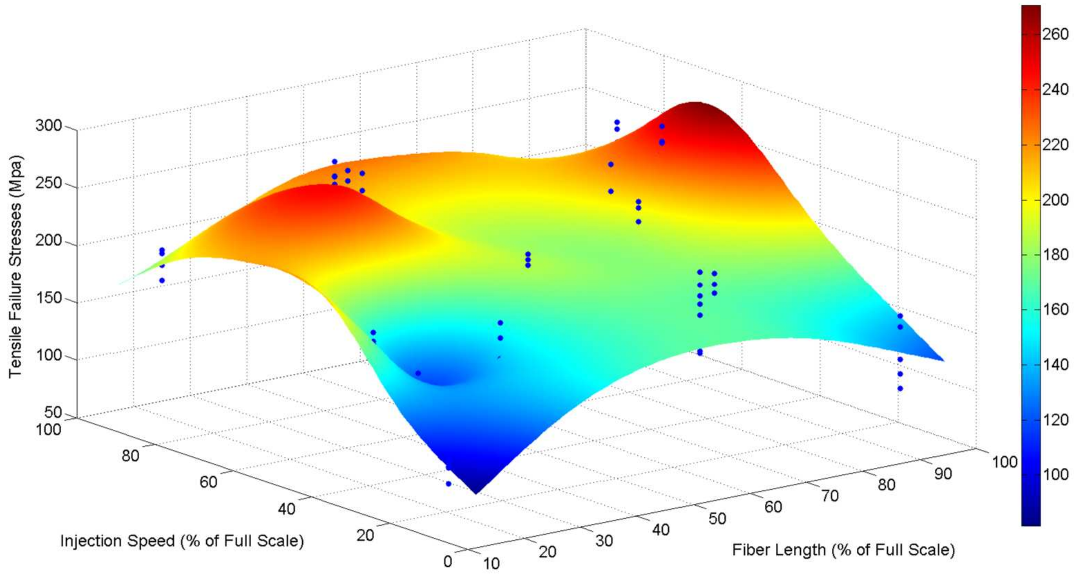


Figure 3. Influence of injection speed and fiber length over tensile failure stress of DCFRC samples manufactured by injection-transfer process

We can clearly remark that σ^R varies from 100MPa to 250MPa whereas results presented Figure 2.b are only included between 100MPa and 150MPa. Failure stresses obviously depend on the four manufacturing paramaters as the surface response demonstrates and not only on fiber length. The surface has a kind of hyperbolic paraboloid shape due to the influence of holding pressure and viscosity of the resin that are not represented in this graphic. Multilinear regression has been done in order to determine the equation linking σ^R to the four manufacturing parameters. The same work has been done for the failure strain ε^R and the elasticity modulus E. Thus, the influence of each manufacturing parameter on tensile properties can be determined. These equations are confidential but the Table 1. presents the effect of an increase in each manufacturing parameter of the injection-transfer process on tensile properties.

	Modulus of elasticity (E)	**Fracture stress (σR)**	**Fracture Strain (εR)**
Fiber length (mm)	↑ (increase)	↑	≈ ↑ (poor increase)
Injection speed (mm/s)	↑	↑	↑
Injection temperature (°C)	≈ ↓ (poor decrease)	↓ (decrease)	x
Holding Pressure (bars)	x (no influence)	↓	x

Table 1. Influence of the increase in each manufacturing parameter over tensile properties of a DCFRC material manufactured by injection-transfer

The increase in the injection speed increases all the mechanical properties (σ^R, ε^R, E) during tensile tests. This manufacturing parameter has an influence over the orientation of fibers as Odenberger et al. [5] observed in the case of the flow of SMC during the closure of the mold. In the injection-transfer process, an increase in the injection speed should force the orientation of fibers in the direction of flow, which is also the load direction during the tensile tests. On the contrary, low injection speed should lead to a more isotropic orientation state.

The influence of injection speed over the microstructure of the material has been observed using ultrasonic C-scan. The results are presented Figure 4.

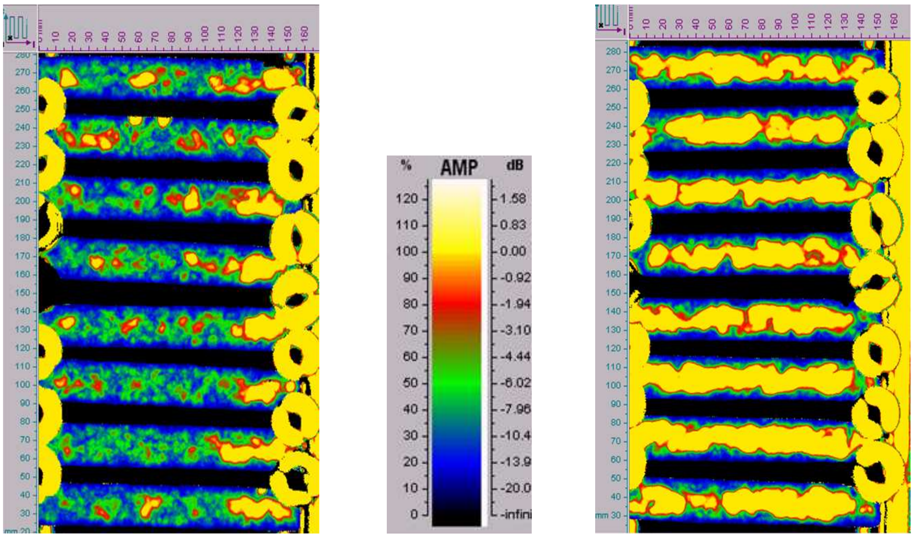


Figure 4. Observation of the influence of injection speed used for manufacturing discontinuous fibers injected samples over the ultrasonic's smapping shape : a) low injection speed – b) high injection speed

The two series of eigth samples analysed on Figures 4.a and 4.b have the same fiber length (17% F.S.). The samples Figure 4.a have been manufactured using a very slow injection speed (15% of the full scale), whereas those on Figure 4.b were manufactured using a high injection speed (100% of the full scale). The average failure stress obtained during tensile tests for the low velocity samples was 97MPa and the elasticity modulus was 33GPa. The samples manufactured with a high injection speed had an average failure stress of 171MPa and an elastacity modulus of 39GPa.

Yellow areas correspond with fields of the material where the amplitude of the ultrasonic waves detected by the sensor correspond with 100% of the ultrasonic signal sent. On the contrary, black areas correspond with fields of the material where no signal is detected. It appears that samples manufactured with low and high injection speed have significantly different ultrasonic mapping. However, they have the same fiber volume rate (56%) and void volume rate (<2%). These values have been determined after chemical matrix digestion of composites samples. So high void volume rates and local resin excess can not explain these differences in ultrasonic mappings. The orientation of fibers should be the explanation : high injection speed orientates fibers in the direction of flow during the filling of the mold. 100% of the ultrasonic wave amplitude is received in this case. On the contrary, low injection speed leads to a more isotropic orientation which involve multi-reflection of the ultrasonic waves. This should be the reason why the amplitude of the received signal is very low in this case. It is consistent with the fact that mechanical properties in the X-direction are lower in the case of low injection speed than for high injection speed.

Damage occurring on this material during tensile tests was also studied. Several load-unload cycles were applied to the rectangular samples. The load steps consisted in increasing progressively the strain in the inelastic area until the fracture of the material. Few cycles have to be applied in order to limit the fatigue behavior of the material. The curve Figure 5 shows an example of load-unload cycles.

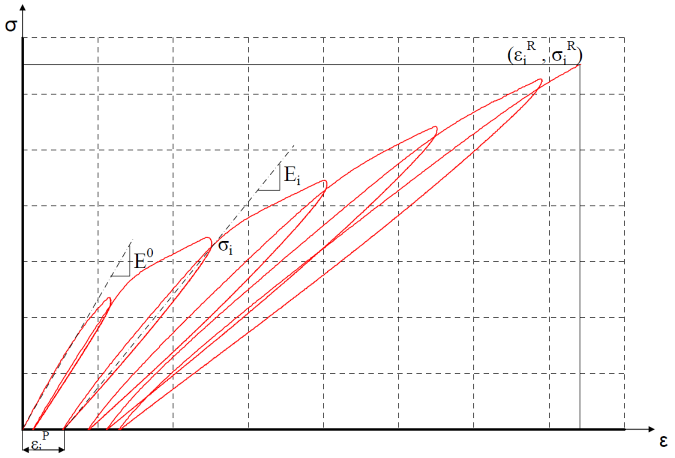


Figure 5. Load-unload cycles for damage study during tensile tests

For each load-unload cycle, the modulus of elasticity of the material decreases which means that the material damages. E_0 is the initial modulus of elasticity (before any damage) while E_i are the damaged modulus calculated on each load. σ_i is the maximal stress during each i cycle, and ε_i^P is the permanent strain that appears and increases when the material damages. Traditionally diffuse damage is characterized by studying the damage variable evolution law. Damage variable d_i must be plotted according to total damage Y_i [6].

The damage variable d_i has been computed for each cycle using the equation 1:

$$\boldsymbol{di = 1 - \frac{Ei}{E0}} \tag{1}$$

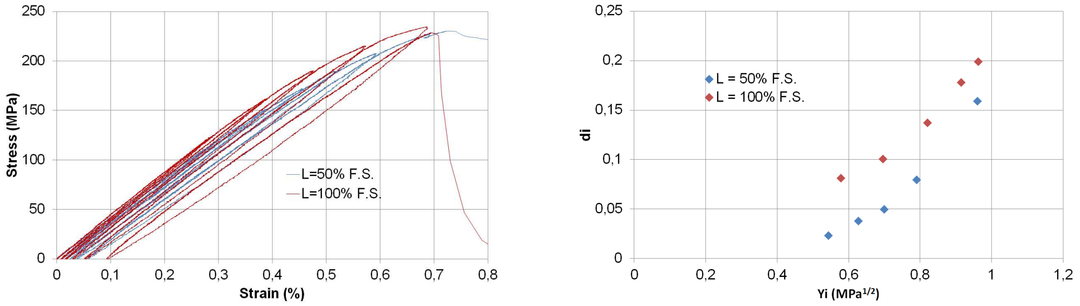
Then the energy release rate Yd_i was computed using equation 2:

$$\boldsymbol{Ydi = \frac{1}{2}\frac{\sigma i^2}{E0(1-di)^2}} \tag{2}$$

The total damage Y_i can finally be computed using the equation 3:

$$\boldsymbol{Yi = \sqrt{Ydi}} \tag{3}$$

Damage of the material manufactured by injection-transfer process was studied for different fiber length. Figure 6.a represents load-unload cycles of two samples manufactured with the same manufacturing parameters. The only difference is the fiber length: 50% F.S. for the first and 100% F.S. for the second. Figure 6.b represents the evolution of damage law $d_i=f(Y_i)$ for these two fiber lengths.

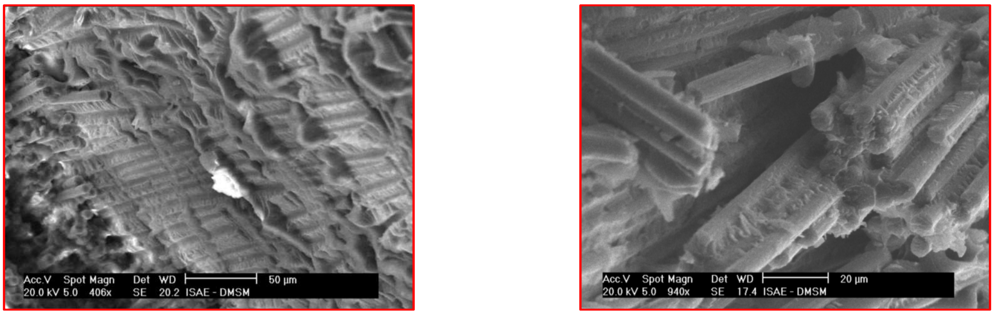


a) b)

Figure 6. Influence of fiber length over tensile damage of DCFRC material manufactured by injection-transfer : a) load-unload cycle – b) di=f(Yi) damage evolution law

It seems that permanent strains of the 100% F.S. fiber length material are more significant than for the 50% F.S.. It would mean that increasing the fiber length would lead to higher damage. This is confirmed by the evolution of damage law $d_i=f(Y_i)$ for the two fiber lengths. The damage variable corresponding with a specific stress level or with a specific Yi is higher in the case of 100% F.S. fiber length material. It means that the higher the fiber length is, the higher damage is. Nervertheless, the maximum damage variables reached with this material are low compared to those reached for other discontinuous fibers composites as SMC-R [7] material.

The main fracture mode has been identified with observation of fracture facies using SEM. Pictures are presented Figure 7.



a) b)

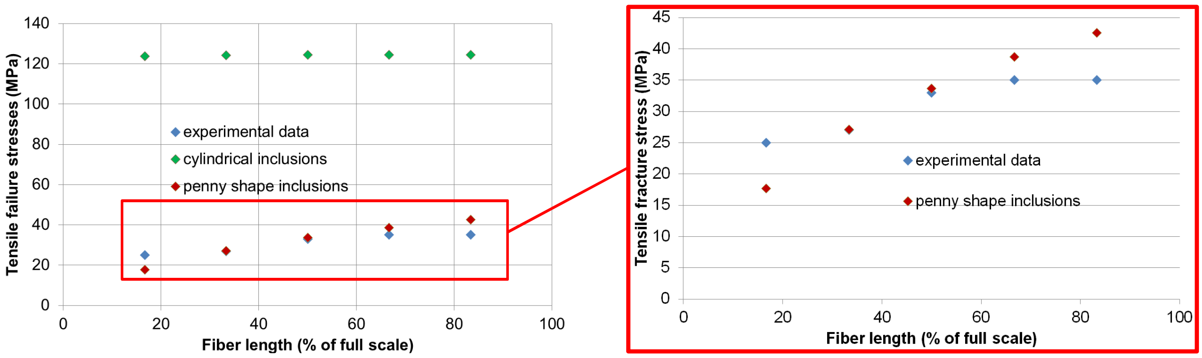
Figure 7. Tensile fracture facies of DCFRC material manufactured by injection-transfer : a) debonding of some chips of fibers from the others – b) debonding matrix/fibers

The marks of chips of fibres can be observed on the resin on Figure 7.a. It is the evidence of the debonding of some chips of fibres from the others. Figure 7.b shows several fibers covered with very few matrix or even without any matrix. The fracture mode is debonding of matrix from fibers in that case. Very few fiber fractures can be observed. As debonding of chips of fibres from the others and debonding of matrix from fibres are the main fracture modes, an increase in the fibre length amounts to increase the covering surface. This is the reason why the higher the fibre length is, the higher maximum damage is.

3.2 Numerical model for prediction of mechanical properties of DCFRC

A numerical model has been developed in order to predict the elasticity modulus of DCFRC manufactured by injection-transfer. It is a micromechanical model which uses Mori-Tanaka's method [8]. The composite material is considered as a matrix containing several inclusions (the fibers). The model computes the elasticity modulus of the composite using the properties of the matrix without any inclusions, the properties of each inclusion and the orientation of the inclusions.

Two kinds of inclusions are tested. The first is cylindrical inclusion which represents fibres as cylinders. Their aspect ratio r=L/Ø is needed, where L is the fiber length and Ø the diameter of the fiber. That kind of inclusion was used to predict the elasticity modulus of short glass fibres and thermoplastic matrix injected composite manufactured by injection [2]. The aspect ratio was below 20. For fibers longer than 1mm, the aspect ratio is always above 140 which is very high. The second sort of inclusion is the penny shape [9]. It represents chips of fibres as a disc. The inclusions are not considered as single fibres but as chips of fibres which corresponds more with the injection transfer-process studied. Penny shape's diameter corresponds with the fiber length of the fibers located in the chips of prepreg that are injected in the mold and that constitute the final composite part. The simplifying assumption made is that fibers located in the same chip of prepreg are not separated during the filling of the mold. The results of the model predictions for the two kinds of inclusions are presented Figure 8 according to fibre length. Experimental data are also plotted.



a) b)

Figure 8. Prediction of the elasticity modulus of a medium length carbon fibers and epoxy resin material manufactured by injection-transfer on: a) full scale results – b) focus on penny shape inclusions results

Cylindrical inclusions always overestimate the elasticity modulus whereas penny shape inclusions leads to better predictions, particularly for 33% F.S. and 50% F.S. fibre length. In these cases, the shape of the inclusions used in the model (red circles in Figure 9) is more conform to those of the inclusion really contained in the composite material (blue rectangles).

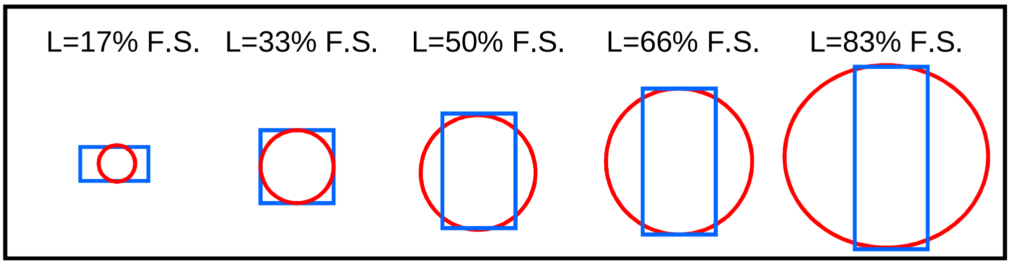


Figure 9. Comparison between inclusions shape used in the model and inclusions used to manufacture the material for each fiber length studied

In order improve the prediction of the model plane-parallel inclusions will be tested since it is more conform to the chips of fibers injected during the manufacturing of parts.

Conclusion

The influence of the manufacturing parameters of the injection-transfer process over tensile properties has been determined. The two main parameters are injection speed and fiber length. Their increase leads to an increase in tensile properties. An increase in the injection speed orientates the fibers in the flow direction during the mold filling. Isotropic fibers orientation seems to be detectable using ultrasonic scans. That point will be confirmed by studying the elasticity modulus of the material in several directions.

This material does not damage a lot. Nevertheless, an increase in fiber length increases the damage.

Homogenisation method using penny shape inclusions leads to promising predictions. Plane-parallel inclusions will be tested. Better results are expected.

References

[1] Vincent M., *Etude de l'orientation des fibres de verre courtes lors de la mise en œuvre de thermoplastiques chargés*. Thèse de l'école Nationale Supérieure des Mines de Paris (1984).

[2] Haramburu E., *Approche intégrée du dimensionnement mécanique de structures en composite injecté avec fibres courtes : une interface entre injection et calcul de structure*. Thèse de l'Université Paul Sabatier de Toulouse III, (2003).

[3] Dray Bensahkoun D., *Prédiction des propriétés thermo-élastiques d'un composite injecté et chargé de fibres courtes*. Thèse de l'Ecole Nationale Supérieure d'Arts et Métiers, (2006).

[4] Blanc R., *Etude de l'injection de composites polyesters thermodurcissables*. Thèse de l'Ecole des Mines de Paris, (1988).

[5] Odenberger P. T., Andersson H. M., Lundström T. S. Experimental flow-front visualisation in compression moulding of SMC. *Composites: Part A*, 35, pp. 1125–1134 (2004).

[6] Ladevèze P. Le Dantec E. Damage modeling of the elmentary ply for laminated composites. *Composites Science and Technology*, 43, pp. 257-267 (1992).

[7] Al-Magrihbi A., *Comportement des matériaux composites à fibres courtes : applications à l'impact basse vitesse*. Thèse de l'université Paul Sabatier de Toulouse III (2007)

[8] Mori T., Tanaka K. Average stress in matrix and average elastic energy of materials with misfitting inclusions. *Acta Metallurgica*, 21, pp. 571-574 (1973).

[9] Mura T. *Micromechanics of Defects in Solids*. Kluwer Academic Publishers, Dordrecht, Boston, London (1987).