PROCESSING-DEPENDENT MICROSTRUCTURE OF LONG GLASS FIBRE REINFORCED POLYAMIDE 6-6 INJECTION MOULDINGS AND RELATED MECHANICAL PROPERTIES

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ABSTRACT: This paper aims at identifying the main parameters that govern the flexural properties of long glass fibre / polyamide 66 injection-moulded parts. The mould geometry has been chosen so as to reproduce some geometrical accidents (e.g. sharp frontal and tangential steps) occurring on industrial moulds. A Taguchi Design of Experiments (DOE) analysis has been devised in order to quantify the processing conditions effects on the flexural strength and modulus. The polymer melt temperature is the main parameter acting on the flexural properties in both flow and transverse directions. The structure/process/flexural properties relationship has then been deduced from a microstructural analysis (local residual fibre length and average orientation, interfacial quality). For optimised injection moulding conditions, leading to the highest flexural strength in the flow direction, a fibre content gradient has been noticed over the part length and width, which is strongly amplified by the presence of a sharp geometrical discontinuity.

KEYWORDS: Long glass fibre, Injection moulding, Microstructure, Polyamide 66

INTRODUCTION

The use of short fibre reinforced thermoplastics in industrial applications is frequently hindered by their rather low mechanical performances, because of the low residual fibre length after processing. In order to overcome this limitation, long fibre reinforced thermoplastics (LFT) have been developed to bring better mechanical properties and to meet the market demand thanks to an improvement of the reinforcement efficiency. At the present time, LFT is one of the fastest-growing sectors of the plastics industry, especially in business areas where high mechanical performances and time stability are required. The automotive applications currently account for over 95% of the worldwide demand. In Europe, an annual growth rate of 10-12% was observed during the 1999-2002 period and car makers forecast further annual growth perspectives of 10% until 2010 for glass reinforced thermoplastics [1,2]. Considering the constraints that such engineering parts have to satisfy, it has become crucial to gain an accurate knowledge of the processing conditions / microstructure / part properties relationship of injection moulded LFT, so as to be able to further optimise their use

potential. However most of the studies on processing-induced fibre orientation and degradation mechanisms carried out up to now were dedicated to short glass fibre injection moulded thermoplastics. Few data is available for LFT injection moulding [3-8], and, in that case, investigations have been usually carried out on very simple (plane) parts, mostly on PP matrices.
As a consequence, this paper aims at contributing to an understanding of how the processing conditions affects the anisotropy and heterogeneity of long glass fibre / polyamide 6-6. An analysis based on a detailed investigation of the injection moulding process at the different elementary stages of plastication and flow will be carried out for this purpose. It will lead to the identification and subsequent optimisation of the main parameters that improve the flexural properties of PA6-6 LFT injection moulded parts.

EXPERIMENTAL

The experiments were carried out on a 2000 kN clamping force injection moulding machine (DK Codim). The machine had an injection gate located in the parting line and a standard 55 mm diameter screw. The prototype injection mould was a rectangular plate of 300 x 120 x 3 mm. The feeding of the cavity was made by a 4 mm thick fan gate over its whole width (unidirectional flow in the longitudinal direction of the plate). This mould was specially designed so as to reproduce some geometrical discontinuities (like frontal and tangential steps) occurring on industrial moulds and to be able to study the effects of such more or less sharp accidents on the flow mechanisms and related part properties (Fig. 1).

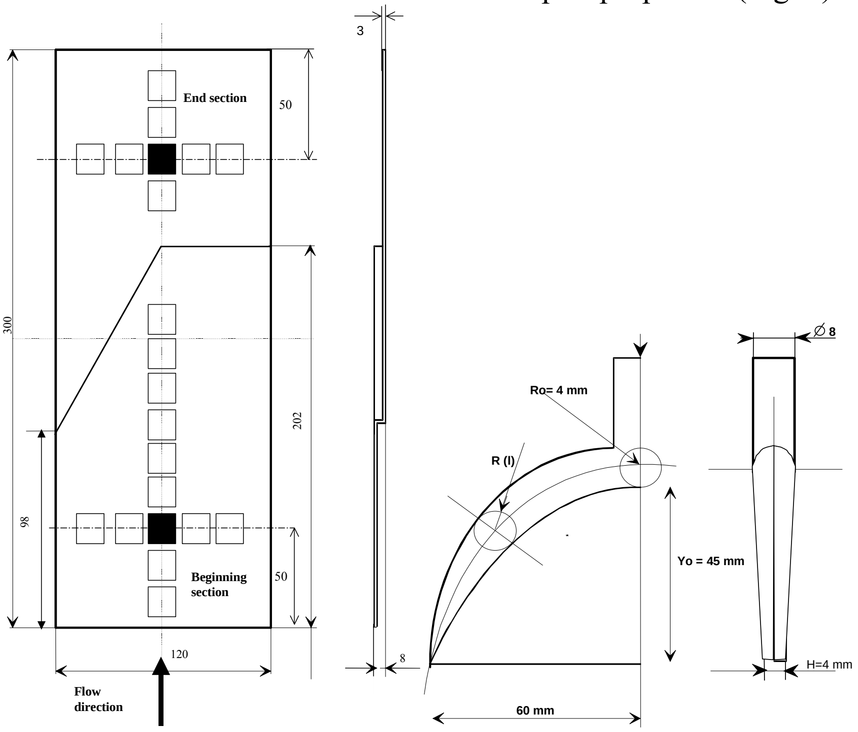

Fig.1: Plastic part and fan gate geometry and cutting pattern of test samples for fibre content (white coupons) and fibre length and orientation (black coupons) determination

A Design Of Experiments (DOE) analysis was devised to identify the main parameters inducing anisotropy and heterogeneity of the moulded part, and then to optimise the injection moulding process of a polyamide 66 (PA66) reinforced by 40 wt% of 10 mm long glass fibres (Ticona). The Taguchi L16 (2^{15}) table used included six factors related to the filling, holding and plastication stages (Table 1) and six interactions between factors. The level of each factor was determined from the material supplier's data sheets. The others processing parameters (holding time, packing pressure, cooling time …) were kept constant. The output parameters were the flexural mechanical properties (strength, defined as the maximum stress, and modulus). Bending tests were performed according to ISO 178 on a standard tensile machine (Instron) on 5 samples (dimensions 60x25x3 mm) in both flow (longitudinal) and transverse directions at the beginning and at the end of the part according to figure 1.

Table 1. Processing conditions for Taguchi DOE

Parameters (or factors)	**Low Limit**	**High Limit**
Mould Temperature (°C) **MoT**	90	120
Melt Temperature (°C) **MeT**	280	300
Volume flow rate (cm^3/sec) **IS**	83	142
Holding Pressure (Bar) **HP**	277	440
Back Pressure (Bar) **BP**	8	12
Screw rotation speed (cm/min) **SRS**	691	1036

For the fibre length measurements, three 25x25 mm size samples were cut from three injection moulded plates (Fig.1) and then burnt at 530°C during 5 hours. The burning residue was scattered in water by ultrasounds and then dried. The remaining fibres were dropped on a glass slide. A polarizing microscope (Jenapol, CarlZeiss Jena) was used in transmission mode and associated to a CCD camera coupled to a computer to get and record suitable images. These images were then analysed by means of an image processing software package (Visilog® 5.2, Noesis). The analysis of 1000 fibres per sample at least led to the determination of the average fibre length and the distribution of fibre length. The number-average fibre length *Ln*, the weight-average fibre length *Lw* and the corresponding standard deviation on the sample were calculated according to ISO 22314 [9]

Finally, the fibre orientation state of the moulded part were describe by the Advani and Tucker [10] method that uses the notion of orientation tensors, in order to obtain a complete description of the orientation state from a small number of discrete values. Such tensors are defined as the dyadic products of the unit vector $\underline{p}$ averaged over all possible directions, with ψ as weighting function. Using an orientation tensor is equivalent to approximating the orientation distribution function by a finite term number in a Fourier series. The second order tensor $\underline{\underline{a}}_2$, defined by equation 1, was used.

$$\underline{\underline{a}}_2 = a_{ij} = \oint p_i \; p_j \; \psi(\underline{p},t) \, \delta \underline{p} \qquad (1)$$

These tensor values characterize the orientation state with respect to a reference direction (observation direction 1), usually the flow direction ($a_{11}<0.35$ for a perpendicular orientation, $a_{11}>0.7$ for a parallel orientation, $0.5<a_{11}<0.6$ for a random orientation)

MANUFACTURING CONDITIONS EFFECT

The main injection-moulding parameters governing the mechanical properties were identified from a variance analysis [11]. From a general point of view, all parameters that decrease the shear stresses in the melt during plastication and flow increase the mechanical properties [6,7,8,11,12]. Melt and mould temperatures as well as volumetric flow rate thus limit the fibre degradation and increase the part quality whatever the location may be. A strong anisotropy effect was also noticed (properties are on average about 1.3 to 2 times higher in the flow direction than in the transverse one) because of a preferential fibre orientation in the flow

direction. The geometrical discontinuities increase the anisotropy as they increase the shear stresses in the flow and thus the fibre orientations.
In order to establish the processing – mechanical properties – structure relationship, two opposite processing conditions were extracted from the general L16 DOE. The first set of processing conditions was deduced from the experimental and analytical model that leads to the maximum flexural strength in the flow direction. This set up of injection machine is named «maximum combination». The second set of processing conditions was chosen at the opposite experimental levels. It is named «minimum combination». The corresponding machine set-up for these combinations are summarized in table 2.

Table 2. Processing conditions for the Structure/Process/Property analysis

Parameters	**Max. Comb.**	**Min. Comb.**
Mould Temperature (°C) **MoT**	120	90
Melt Temperature (°C) **MeT**	300	280
Volume flow rate (cm^3/sec) **IS**	83	142
Holding Pressure (Bar) **HP**	440	440
Back Pressure (Bar) **BP**	8	12
Screw rotation speed (cm/min) **SRS**	1036	691

MICRO-STRUCTURAL ANALYSIS

Fibre content

The average fibre weight content is 41.5wt% and 42wt% respectively for the minimum and maximum combinations. However, the glass weight content measurement highlights a fibre accumulation at the parts end section for both combinations, with a maximum of 51wt% of fibres noticed for the maximum combination (Fig.2). In the transverse direction, the parts beginning section is homogeneous for both combinations, contrary to the end section where the maximum fibre content is observed on the symmetry axis (Fig.3). Such a concentration gradient was also observed by some authors both for short [13,14] and long fibres [15]. It may be attributed to the sweeping towards the parts end of the fibres, which are partially embedded at the interface between the frozen and molten layers and broken by flow-induced shear stresses.
For the minimum combination, the mould and melt temperatures are set at their lower level and the volume flow rate at its higher level. It is possible to suppose that the shear flow in the skin layers is favoured compared to the extensional flow, leading to a higher longitudinal migration at least in the first half part of the plate (Fig. 2). The fibre orientation profile, measured hereafter, is going to confirm this hypothesis.
For the maximum combination, the filling time is 1s higher and the freeze time 3s higher due to a lower volume flow rate and a higher mould or melt temperature. Even if the shear stresses during the filling stage are lower and the frozen outer layers thinner than for the minimum combination, the flow effect during the holding pressure stage is greater, and this induces a higher fibre concentration gradient. An analysis of the glass content of each layer would be required in order to confirm this hypothesis.

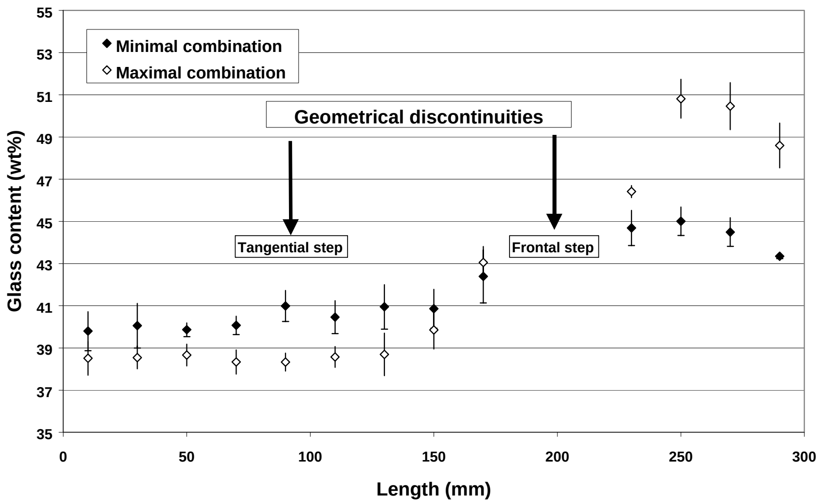

Fig.2: Glass weight content along the mid-axis of the composite plates

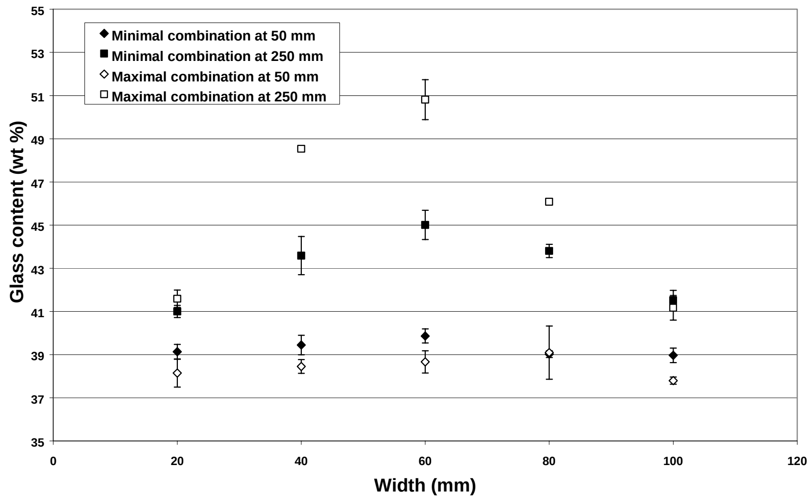

Fig.3: Glass weight content along the width of the composite plates.

Fibre length

The maximisation of the processing conditions allows increasing the whole reinforcement average length (both number- and weight-average) of about 20% (Tab.3.).

Table 3. Fibre length measurements

	Min. comb.		Max. comb.	
	Begin.	***End***	***Begin.***	***End***
Number-average length *Ln* (mm)	1.372	1.262	1.503	1.802
Weight-average length *Lw* (mm)	3.114	2.661	3.183	3.839
Standard deviation (mm)	1.573	1.350	1.590	1.917
Polydispersity ratio *Lw/Ln*	2.27	2.11	2.12	2.13
Fibre aspect ratio	91	84	100	120

For the maximum combination, the number-average fibre length in the parts end section is 20% higher compared to the beginning section. This would reveal fibre degradation from the secondary flows during the holding stage of the process. A lower viscosity of the melt favours the pressure transmission.
An important degradation of the reinforcement could be observed for both combinations. Almost half of the fibres have a length less than 1 mm and only 10% of the fibres are longer than 5 mm. The average degradation amount has been evaluated to 80% of the initial average fibre length.
According to the results presented in table 3, most of the degradation occurs in the cavity up stream, in the plasticating unit or in the feeding system (nozzle or gate). An evaluation of the degradation occurring within the plasticating unit has therefore been carried out for the maximum combination in order to understand the fibre degradation mechanism during processing. It appears that the fibre degradation occurs mainly in the compression section where the degradation amount reaches 70% (Fig.4).

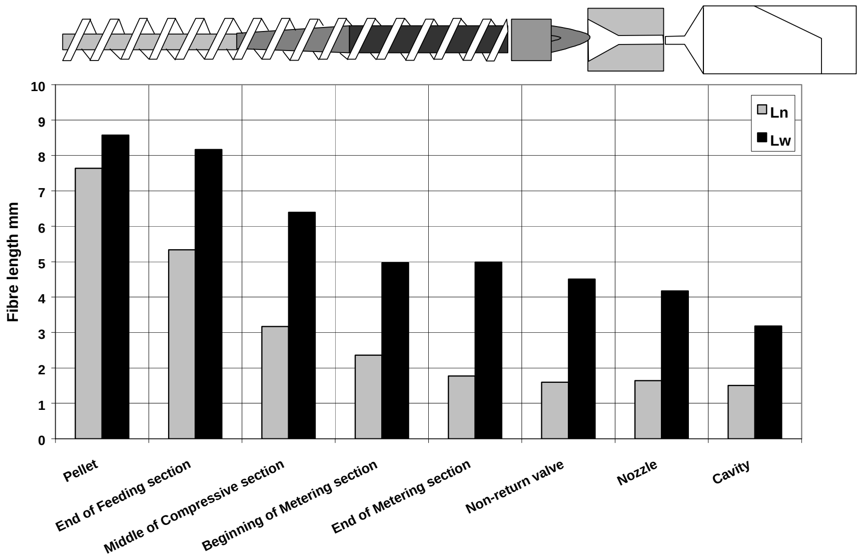

Fig.4: Number- and weight-average fibre lengths along the processing system

Fibre Orientation

The fibre orientations were evaluated as well. The Advani-Tucker [10] tensor values (a11) were plotted through-the-thickness for the two combinations at the parts beginning and end sections (Fig.5).
The orientation profile is symmetric with respect to the plate mid-plane. Seven elementary layers are visible for the maximum combination. There are two skin layers, where the fibres are randomly distributed due to the fountain flow, and two oriented layers due to the shear flow influenced by the injection speed and the non-isotherm effects. Two random intermediate layers resulting from the secondary flows during the holding stage are therefore noticed. Finally, a transversely oriented layer is present in the centre of the parts. The minimum combination shows a 5-layer composite with two thick shear layers. The effect of the holding pressure is similar to that for the maximum combination. At the end of the parts, the a11 tensor value does not change through the thickness due to the sample location on a weld line, which is created there.
The fibre orientation mechanisms seem to be those observed for short fibres at least for fibre length below one millimetre. A the beginning of the cavity, the fibres take a perpendicular orientation in the (1,2) plane due to the divergent flow induced by the fan gate radial

stretching. In the first half of the plate, the flow is unidirectional. The fountain and shear flows influence the fibre orientations as described by GERARD [13]. The geometrical discontinuities disturb the flow and induce a weld line, where the reinforcement is oriented in the flow direction (1).
The thickness of the mould side layer increases from the entry (gate) to the plate end due to cooling. This creates a convergent geometry in the (1,3) plane. This effect is all the more important than the temperature is lower, even for the minimum combination.

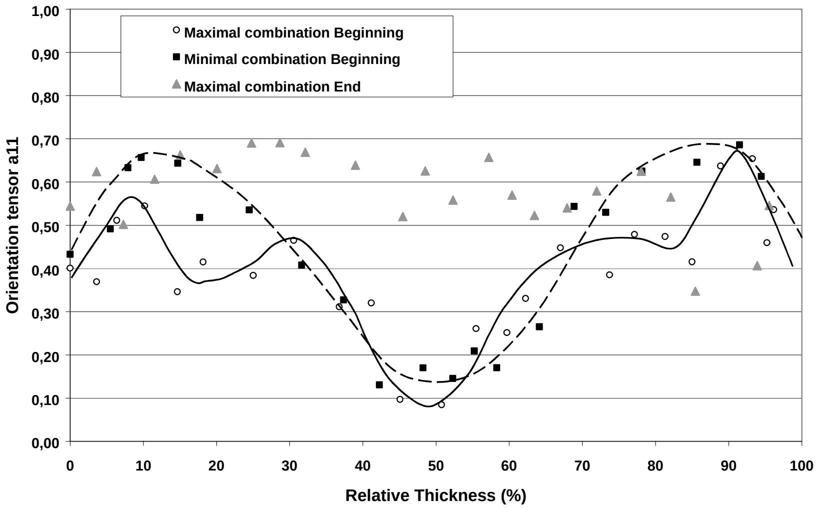

Fig.5: Fibre orientation profiles through-the-thickness of the injection moulded part

Interfacial adhesion evaluation

The quality of the interfacial adhesion were evaluated by observation of the fracture surfaces under Scanning Electronic Microscope (SEM, Phillips). Generally, the samples moulded with the maximum combination are characterised by a better interfacial adhesion. The matrix perfectly covers the fibres and a cohesive fracture is noticed. On the opposite, when the minimum combination is used, the interfacial adhesion quality is poor, the fibres are smooth, the fracture is non cohesive with a matrix/fibre slipping.

CONCLUSIONS

A detailed microstructure analysis of the moulded parts has pointed out some features related to optimised processing conditions:

- ✓ a strong fibre content gradient with higher contents at the end of the parts, which can be attributed to a longitudinal segregation induced by shear stresses in the melt, and is amplified by the presence of sharp geometrical accidents;
- ✓ a higher average fibre length (at least +20% to +40% depending on the location in the part);
- ✓ a 7-layer composite structure with a great effect of the holding pressure.

The analysis of the structure of the parts has also shown that the high flexural properties of the parts moulded with the maximum combination of machine set-up parameters mainly come from the reinforcement content and average length combined to a better interfacial adhesion of the matrix around the fibres.

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