OPTIMISATION OF THE LOW-SHEAR COMPOUNDING OF CARBON NANOFIBRE / POLYMER COMPOSITES

F.W.J. van Hattum[1], A. Gaspar-Cunha[1], M.L. Lake[2], and C.A. Bernardo[1]

[1]IPC – Institute for Polymers and Composites, University of Minho, 4800-058 Guimarães, Portugal
[2]Applied Sciences, Inc., P.O. Box 579, Cedarville, OH 45387, USA

ABSTRACT

During thermoplastic processing, typically high levels of shear are introduced into the polymer material which is beneficial for polymer melting and mixing quality. Composite properties, however, depend largely on the fibre length retained in the material [1], which degrades rapidly with applied shear. The current work describes a methodology to optimise the production of carbon nanofiber thermoplastic composites by balancing applied shear and fibre degradation. The optimisation approach used combines a modelling routine of the plasticating extrusion process with a Multi-Objective Evolutionary Algorithm (MOEA) able to deal with multiple objectives [2,3]. In this work carbon nanofiber nylon masterbatches have been compounded and subsequently injection moulded at several fibre loads. The resulting composite properties are analysed and used as criterions in the processing models. In this way, the operating conditions and/or to design extrusion screws can be optimised taking into account some critical parameters related to the minimisation of fibre degradation during compounding.

1. INTRODUCTION

Thermoplastic polymer composites incorporating carbon nanofibers can combine beneficial levels of mechanical properties with improved transport properties like electrical and thermal conductivity that allow for applications where functions such as EMI-suppression and electrostatic discharge (ESD) are an issue. The level of these properties however depends largely on the carbon nanofiber dispersion in the composite as well as their aspect ratio.

To incorporate reinforcing fibres in a thermoplastic matrix, typically extrusion compounding methods are used. In these processes usually high levels of shear are applied, that benefit the melting of the thermoplastic polymer as well as the mixing quality of the composite. However, the high shear levels involved will, although improving dispersion, at the same time lead to a significant break-up of the fibres, thus having a counter effect on final composite performance. In order to optimise composite performance a trade-off must be sought between level of fibre mixing and fibre break-up during compounding. The current work studies the compounding of thermoplastic carbon nanofiber composites by using and optimising critical process-related parameters affecting composite performance through process simulation and is a work in progress. This methodology can in future be used to optimise processing conditions or screw geometry for carbon nanofiber compounding.

2. EXPERIMENTAL

2.1 Modelling and optimisation

The compounding of carbon nanofiber thermoplastic composites by balancing applied shear and fibre degradation was optimised combining a modelling routine of the plasticating extrusion process with a Multi-Objective Evolutionary Algorithm (MOEA) able to deal with multiple objectives. The aim is to set the operating conditions and/or to design extrusion screws taking into account some critical parameters related to the minimisation of the fibre length reduction.

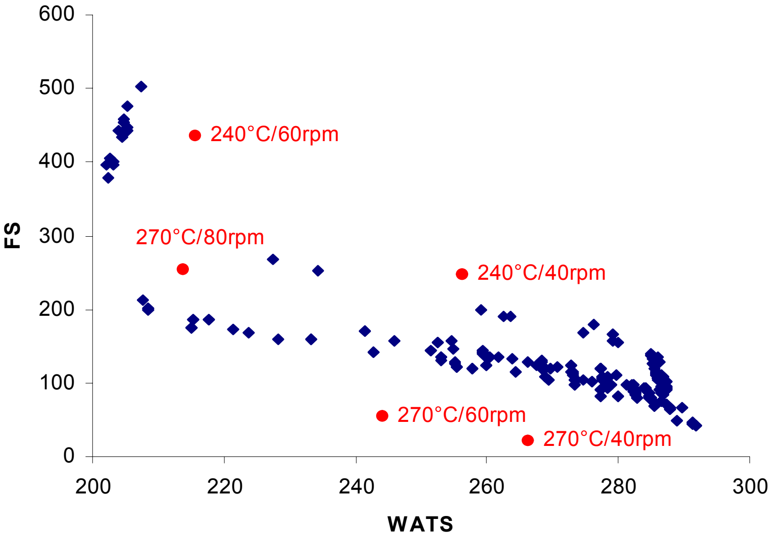
The optimisation approach for the problem is used, that has been successfully applied to several practical problems in single screw extrusion [2,3]. The methodology uses a mathematical model to describe the flow of a polymer in a single screw extruder. This description is used iteratively to define sets of optimised input parameters to reach a desired objective. Two objectives have been defined for optimisation. The Weighted Average Total

Strain (WATS) is an objective related to the total shear deformation undergone by a fibre during processing and should be minimised. The FS (Fibre Shear) is related to the total shear stress that a fibre undergoes in the melt zone, which should also be minimised.

In order to find all possible solutions to the problem based on different sets of input parameters, MOEAs are used in the current work: from a random set of input parameters, the most promising ones, as defined by the output objectives, are combined and used for further generation. This process repeats itself until the best set of input parameters has been identified. The advantage of the use of MOEAs in the current problem is that no solutions are overseen, and only promising solutions are further generated upon.

2.2 Extrusion Compounding

The optimisation algorithm was used for the optimisation of the processing conditions of a Leistritz laboratorial LSM30 single screw extruder, fitted with a ∅36mm screw, with a L/D ratio of 25, a 2.8 compression ratio and a 2mm channel depth in the pumping zone, coupled to a simple cylindrical rod die. As input variables, screw rotation speed and three barrel temperature profiles were considered. Several FS vs. WATS combinations for the current system were calculated using the above scheme, that are plotted in Figure 1.



“**Fig. 1.** FS vs. WATS resulting from the current simulations (red marks indicate input parameter sets used in compounding trials)”

The simulations were aimed at the production of 10 wt% master batches of carbon nanofiber (Pyrograf®-III nanofibers, Applied Sciences), using Nylon 6 (Capron 8202, Honeywell) as a thermoplastic matrix. Compounding trials were run on the Leistritz single screw extruder using different sets of input parameters, based on several combinations of the values of FS and WATS. The parameters were selected based on the extremes of the plot shown in Figure 1, thus giving the most complete description of the relation between different combinations of FS and WATS and ultimate composite properties. The processing parameters used in compounding the several batches of 10 wt% carbon nanofiber composites are given in Table 1.

“**Table 1.** Operating conditions used in compounding trials.”

	Trial				
	1	**2**	**3**	**4**	**5**
Screw speed (rpm)	40	60	40	60	80
Temperature (°C)	240	240	270	270	270

With the conditions used, several master batches were processed. The nylon pellets were dried at 60°C at least 8 h prior to processing, as were the nanofibers, to eliminate humidity. The premixed dry material consisting of nylon pellets with 10 wt% carbon nanofibers was then fed to the extruder. The extruded material was cooled down in a water bath and subsequently cut into pellets. The produced batches were processed on a Klöckner Ferromatik 20 tons injection into test samples using fibre loadings from 0 to 10 wt.% (by diluting with virgin matrix material). The operating condition used are given in Table 2. The mechanical properties have been evaluated by tensile testing the samples in an Instron 4505 Universal testing machine, at a cross-head speed of 2 mm/min. Fibre loadings in the composite materials were determined by density tests.

“**Table 2.** Operating conditions used in injection moulding.”

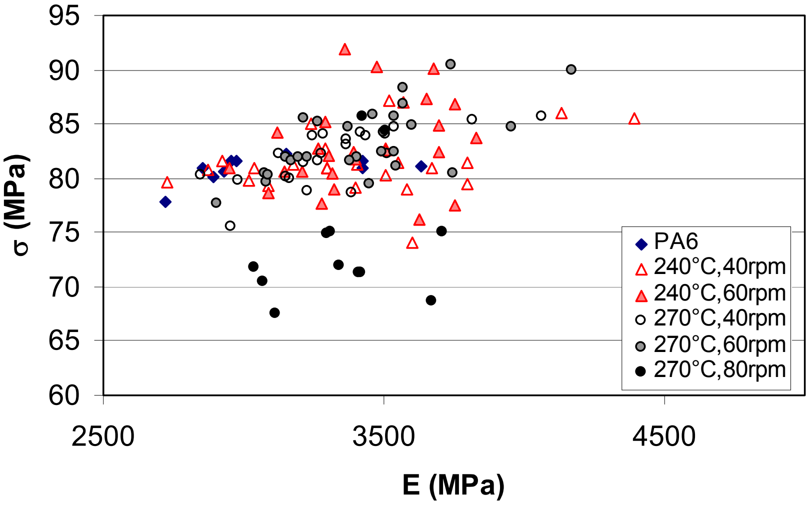
Variable	**Value**
Injection pressure (bar)	80
Holding pressure (bar)	40
Melt temperature (°C)	240
Cycle time (sec)	25

3. RESULTS & DISCUSSION

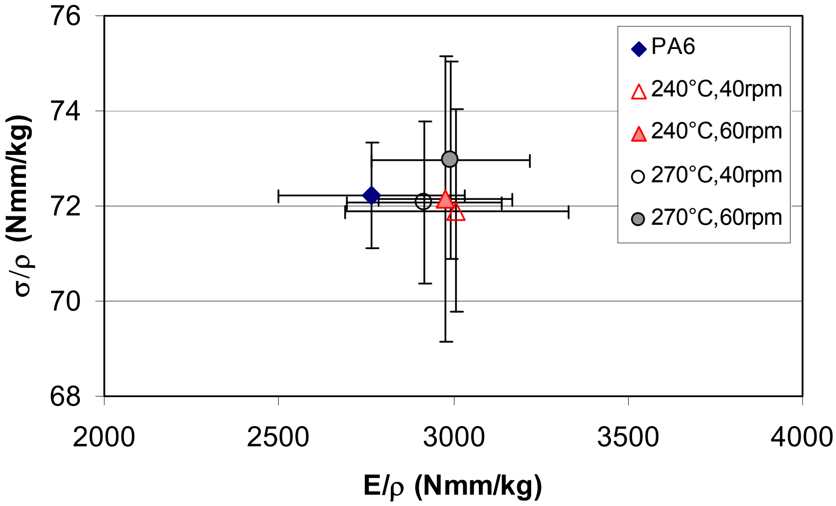
3.1 Mechanical properties

In Figure 2 the strength values obtained on the produced composites are given versus the stiffness. For comparison also the virgin matrix material values have been shown. As can be seen from the figure, higher strength values generally coincide with higher stiffness values, as is to be expected. By far most composite values exceed those of the unreinforced polymer, thus showing an effective reinforcing effect of the nanofibers over the matrix properties. However, as the values in the figure represent a wide range of fibre weight fractions produced, and the observed spread in mechanical properties is relatively small, from the figure it is difficult to accurately observe the differences induced between the different batches produced at different extrusion compounding conditions.

In Figure 3, therefore, the averaged values of the ‘specific’ strength versus stiffness are plotted: the mechanical properties obtained on the samples, normalised by the density of each sample. Thus an indication of the average reinforcing effect per unit of fibre fraction is obtained, allowing an intuitively easy comparison of the different compounding methods used: a value in the top right hand corner of the graph would combine high specific strength with high specific stiffness, whereas values close to the origin indicates poorer reinforcing effect. As can be seen from Figure 3, only marginal differences exist between the different compounding trails: statistical analysis of the results shows that only the values of the compounding trials produced at 270°C and a screw speed of 60 rpm shows a significant improvement over the others, as compared to the matrix material. Please note that in the figure, the results of the trial at 270°C and a screw speed of 80 rpm have been omitted, as the specific strength results fell far outside the range of the others. This effect could already be observed in Figure 2. The exact reason for this behaviour is still under study.

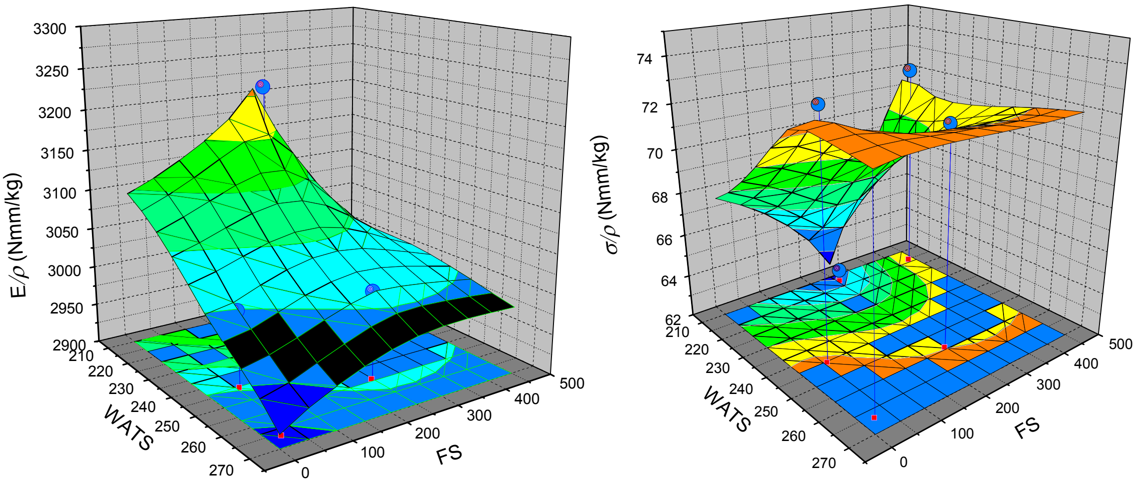


“**Fig. 2.** Strength vs. stiffness of the produced composites.”



“**Fig. 3.** Specific strength vs. specific stiffness of the produced composites.”

To see the relation between the processing parameters WATS and FS, related to fibre breakage, used to optimise compounding and the final composite properties observed, in Figures 4a and 4b the specific stiffness and strength, respectively, have been plotted versus the calculated values of WATS and FS. Although the overall effect of WATS and FS is somewhat distorted by the effect of one compounding trial (the surfaces ‘drawn’ to peak values at these points), the figures show the tendency of higher specific properties being related to intermediate values of WATS as well as low values of FS. Statistical analyses of the data confirm this tendency, although again only for the processing conditions of 270°C with a screw speed of 60 rpm, a statistical significant difference is found. On the one hand side this indicates the robustness of the compounding system used in the current work: changing processing parameters has little effect on final composite properties. At the same time however, herein an overruling effect of the high shear-levels induced by the injection moulding process (usually some decades higher than in the extrusion process) can be reflected. This effect, however, is a real one as in practice the produced compounds will undergo these high shear levels through subsequent processing by injection moulding.



“**Fig. 4.** Specific stiffness a) left, and specific strength b) right, vs. FS and WATS.”

4. CONCLUSIONS

In order to optimise the properties of carbon nanofiber thermoplastic composites, a method has been used to tailor levels of fibre breakage and fibre dispersion in extrusion compounding. Two parameters related to fibre breakage have been used in single screw extrusion simulation, and related to final injection moulded composite performance. Although the produced composites show an increase in mechanical properties by the introduction of the carbon nanofibers, the differences between the effect of the different compounding conditions on the mechanical properties are statistically significant for only one processing condition. This indicates the robustness of the current system, but at the same time could show an overruling effect of the (higher) shear-levels induced by the injection moulding process used. Current evaluation of composite transport properties, that are more sensitive to differences in fibre break-up, as well as studies directly on the compounded material as-produced will this clarify this issue. If proven successful, the current method allows for further optimisation of screw geometry as well as different compounding methods, in order to obtain carbon nanofiber composites with improved performance.

ACKNOWLEDGEMENTS

This work was financially supported by the FCT – Portuguese Foundation for Science and Technology, through the POCTI and FEDER programs. One of the authors (FVH) wishes to acknowledge the support of the Luso-American Development Foundation (FLAD) in funding his travel to the partner site that enabled the activities described in this work.

References

1. **van Hattum, F.W.J.** and **Bernardo, C.A.**, “A Model to Predict The Strength of Short Fiber Composites” *Polym. Compos.*, **20**/4 (1999), 524.
2. **Covas, J.A., Gaspar-Cunha, A.** and **Oliveira, P.**, ”An Optimization Approach to Practical Problems in Plasticating Single Screw Extrusion”, *Polym. Eng. and Sci.*, **39**, (1999), 443.
3. **Gaspar-Cunha, A.** and **Covas, J.A.**, ”The Design of Extrusion Screws: An Optimisation Approach”, *Int. Polym. Process.*, **16**, (2001), 229.