EFFECT OF THE PROCESSING CONDITIONS AND MALEATION ON THE PROPERTIES OF BASALT FIBRE REINFORCED POLYPROPYLENE

J. Kano-Ibarretxe[1], J. Anakabe[1*], M. Iturrondobeitia[2], R. Hernandez[1], A. Arrillaga[1]

[1]*Leartiker, Materials dept., Lea Artibai Ikastetxea S. Coop, Markina-Xemein (Spain)*
[2]*Escuela Universitaria de Ingeniería Técnica Industrial de Bilbao, University of the Basque Country, Bilbao (Spain)*
**janakabe@leartik.com*

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Abstract
The present work aims to compare polypropylene based composites reinforced with basalt and glass fibre of the same tex and diameter but different sizing. Maleic anhydride grafted PP was added to improve fibre-matrix interphase. Processing has been carried out by twin-screw extrusion (Brabender, Ø20, L/D=40) and injection moulding (Demag, 100Tn). In order to determine whether those basalt fibre reinforced PP compounds could become a technically feasible alternative for glass fibre reinforced PP compounds, the influence of the processing parameters on the fibre length and microstructure of the composites have been studied. PP+PP-g-MA/BF composites show better overall mechanical behaviour than the other composites, including PP+PP-g-MA/GF samples. The incorporation of 30% in weight of GF or BF increases the elastic modulus of the matrix up to 7 GPa. Samples with PP-g-MA and 30% of BF or GF show up to 45% improvement on tensile strength and impact resistance.

1 Introduction

The use of thermoplastic matrices in short-fibre reinforced polymer composites (SFRPC) offers important advantages over thermoset matrix polymer composites regarding to processing, handling and recycling. The possibility of higher production rates and lower manufacturing costs had lead to an intensive research work on this kind of composite materials. The key factors to manufacture high mechanical performance SFRPC lie on the equilibrium between the fibre content and their aspect ratio (length/diameter), and the adhesion level between the constituents (interphase). It has been demonstrated that the tensile, flexural and impact properties improve significantly when fibre concentration and/or fibre length are increased [1,2,3]. Generally, the manufacturing processes of SFRPC use to cause high shear on the material resulting on breakage of the fibres due to fibre-matrix, fibre-fibre and fibre-machine walls interactions [4]. Thus, the higher the fibre content, the higher their breakage during processing. This makes difficult to control the influence of both, fibre content and aspect ratio, on the final composite properties.

The other important factor is the behaviour of the interphase between the reinforcement and the matrix, which actually is one of the main influencing factors for every kind of composites.

An effective load transfer through the interphase is essential in order to obtain a real improvement on the properties of the matrix. It is accepted that weak interphases give rise to relatively low strength and stiffness but highly tough composites; while strong interfaces lead to high strength and stiffness but brittle composites [5]. According with the literature [6,7], an efficient method to enhance the adhesion between different types of fibres and polypropylene matrices, and therefore improve the final overall mechanical characteristics of the composite, is the addition of polypropylene grafted maleic anhydride (PP-g-MA) to the system.

The aim of the present work is to compare tensile and impact properties of composites based on a polypropylene matrix and reinforced with basalt and glass fibres. Basalt fibres, due to their exceptional mechanical, thermal and chemical properties, are considered the material to fill the gap between glass and carbon fibres [8]. Hence, the interest in these reinforcing materials which show potential to replace glass fibre in some of its applications. The manufacturing of SFRPC was carried out by extrusion compounding and injection moulding, which are two of the most known industrial processes for thermoplastics [9,10]. In order to conclusively determine whether those basalt fibre reinforced PP compounds could become technically feasible for applications where glass fibre is used exclusively, it is necessary to identify how extrusion and injection parameters affect on the fibre size and microstructure of the different reinforcements and therefore on the final properties of the composite. Besides, the influence of the PP-g-MA incorporation into both systems as coupling agent has been studied, too. This modified polypropylene was added together with the PP matrix in the compounding step and the reinforcement (roving) was fed into the extruder as a continuous fibre strand form in order to obtain the longest fibre length after extrusion. This method should give place to pellets which have fibre lengths over the critical length necessary to reinforce the matrix. The measurement of the fibre length in the pellets was carried out by optical microscopy. Finally, the influence of the PP-g-MA on the adhesion level of the fibre/matrix system was studied by scanning electron microscopy.

2 Materials, processing and characterization

Polypropylene (HK060AE, from Borealis) was used as matrix for all compounds. Basalt fibre (BF) roving (KVT2400Tex17E-I) was obtained from Basaltex-Flocart NV (Belgium) and glass fibre (GF) roving (SE4121 Direct Roving) from 3B-the Fibreglass Company (Belgium). Maleic anhydride grafted PP (Exxelor PO 1020, by ExxonMobil) was used as coupling agent.

Sample	PP	PP-g-MA	Glass Fibre	Basalt Fibre
1	70	-	30	-
2	70	-	-	30
3	56	14	30	-
4	56	14	-	30

Table 1. Weight fractions (percentage) of components for each sample.

All samples (see Table 1) were reinforced with a 30% of fibre content. The organic matrix (70% in weight of the final blend) for samples 3 and 4 was prepared by 80% neat PP and 20% of PP-g-MA by weight (which stand for 56% and 14% in the final blend, respectively).

Compounded samples were prepared by melt blending in a Brabender DSE 20/40D twin screw extruder, then pelletized and finally injection moulded in a full electric Demag IntElect

100t (see Figure 1). Initial and compounded pellets were dried for 2h at 80ºC before extrusion and injection, respectively.

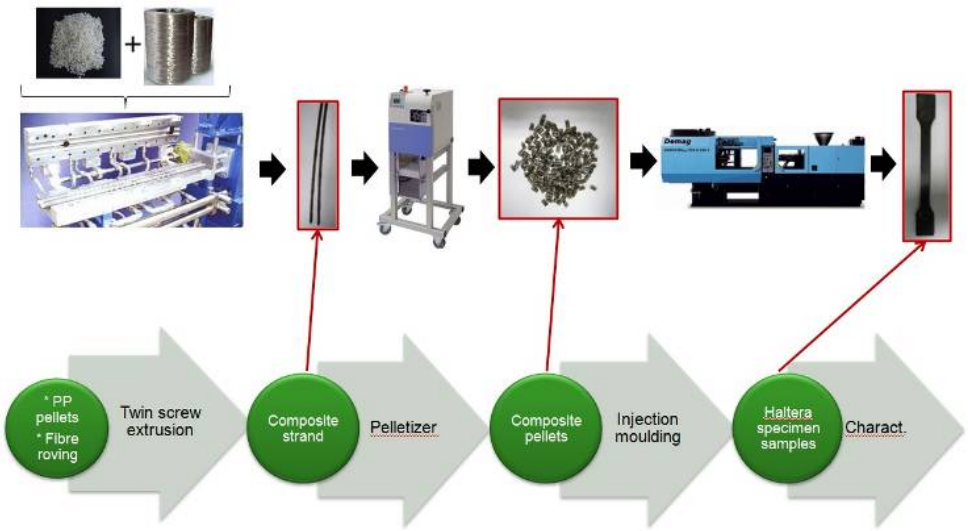

Figure 1. Scheme of the processing steps to prepare the compounds and injection moulding of specimens.

Each compounded sample (see Table 1) was injection moulded at 4 different parameter settings. Variations were high and low plasticization speeds (ω=500rpm, $t_{plasticization}$=2.5s; ω=100rpm, $t_{plasticization}$=15s) and high and low injection speeds (v=160mm/s, $t_{injection}$=0.3s; v=40mm/s, $t_{injection}$=1.2s)

Tensile strength and deformation were measured by a Hounsfield H25KS universal machine at a 10mm/min deformation speed. Young's modulus was measured by a MTS 819 testing system with axial extensometer at a 1mm/min deformation speed. Impact tests were carried out in a ATS impact15 pendulum system (notched Charpy, ISO 179).

Images of fibres after calcination (1h, 625ºC) were obtained by a NIKON SMZ745T stereoscopic microscope. Pre-pelletized (extruded strand) samples, pellets and injected specimens were burned, in order to compare fibre lengths before and after every processing step. From 300 to 600 fibres were measured for each sample.

Hitachi S-4800 scanning electron microscope was used to obtain micrographs of tensile tested breakage surfaces.

3 Results and discussion

3.1 Mechanical properties

Tensile strength and maximum deformation results for composites obtained by each injection parameter setting can be seen in Table 2.

Sample		Tensile strength (Mpa) and maximum deformation (%)									
		ω↓+V↓		ω ↓+V↑		ω ↑+V↓		ω ↑+V↑		Average	
		σ (MPa)	ε (%)	σ (MPa)	ε (%)	σ (MPa)	ε (%)	σ (MPa)	ε (%)	σ (MPa)	ε (%)
PP/GF	Average	50,3	1,50	50,7	1,53	49,5	1,61	49,3	1,66	50,0	1,57
	St. Dev.	*0,8*	*0,06*	*0,9*	*0,06*	*0,9*	*0,08*	*0,2*	*0,05*	*0,7*	*0,07*
PP/BF	Average	51,0	1,47	54,8	1,61	51,4	1,52	51,4	1,57	52,2	1,54
	St. Dev.	*3,2*	*0,13*	*1,2*	*0,10*	*0,6*	*0,06*	*0,6*	*0,04*	*1,8*	*0,06*
PP+PP-g-MA/GF	Average	86,5	2,96	84,9	2,93	83,3	2,90	81,7	2,99	84,1	2,94
	St. Dev.	*0,2*	*0,08*	*0,6*	*0,10*	*0,3*	*0,02*	*0,5*	*0,04*	*2,1*	*0,04*
PP+PP-g-MA/BF	Average	93,4	3,39	91,8	3,39	91,8	3,30	90,3	3,37	91,8	3,36
	St. Dev.	*0,7*	*0,11*	*0,2*	*0,06*	*0,2*	*0,11*	*0,5*	*0,08*	*1,3*	*0,04*

Table 2. Tensile strength and maximum deformation of injected specimens.

Surprisingly it seems that injection conditions have little influence on the obtained mechanical properties (see Figure 2). This indicates that the length of the fibres in the composites might be very similar in all cases.

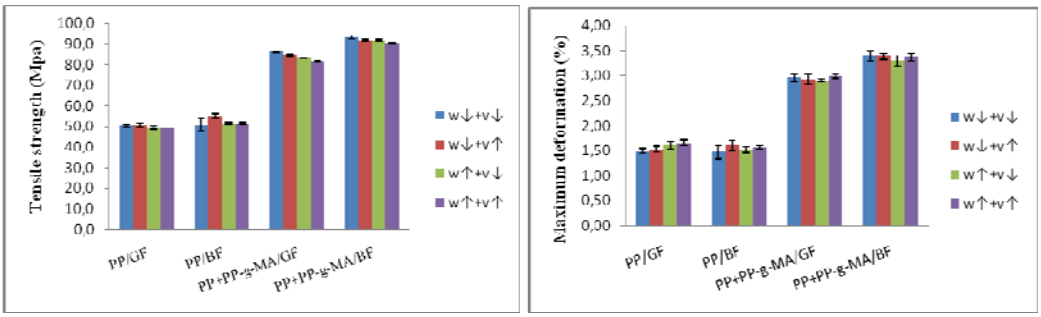

Figure 2. Mechanical properties. Little influence of the injection parameters is shown.

BF shows similar reinforcing efficiency to glass fibre, which makes it a suitable alternative for GF reinforced products. In the other hand, for both BF and GF, tensile strength and maximum deformation are greatly improved by the addition of maleic anhydride grafted PP to the composite. Tensile strength is improved 68% for GF composites and 76% for BF composites; and maximum deformation 87% for GF and 120% for BF. This means that the use of a coupling agent when compounding this type of composites is necessary to achieve interesting mechanical properties.

Lastly, injection parameters seem to have no major influence on the elastic modulus of the composites (see Table 3). However, slightly better results are obtained when using low plasticising and injection rates.

Sample		Young's Modulus (GPa)				
		ω↓+v↓	ω ↓+v↑	ω ↑+v↓	ω ↑+v↑	Average
PP/GF	Average	6,5	6,3	5,9	6,0	6,2
	St. Dev.	*0,2*	*0,1*	*0,5*	*0,2*	*0,3*
PP/BF	Average	6,7	6,3	6,0	6,4	6,3
	St. Dev.	*0,1*	*0,8*	*0,2*	*0,2*	*0,3*
PP+PP-g-MA/GF	Average	6,8	6,6	6,3	6,5	6,5
	St. Dev.	*0,3*	*0,3*	*0,0*	*0,1*	*0,2*
PP+PP-g-MA/BF	Average	6,9	6,8	6,7	6,4	6,7
	St. Dev.	*0,1*	*0,1*	*0,2*	*0,3*	*0,2*

Table 3. Young modulus of samples injected at different conditions.

Regarding impact resistance, the results displayed in Table 4 show that GF seems to reinforce slightly more than BF, at least for samples without coupling agent. When PP-g-MA is added, GF reinforced composite's resilience is improved 65% (from 7,1 J/m^2 to 11,7 J/m^2) on average, and BF reinforced composites an 83% (from 6.4 J/m^2 to 11.7 J/m^2).

Sample		**Resilience (KJ/m^2)**				
		ω↓+v↓	ω ↓+v↑	ω ↑+v↓	ω ↑+v↑	Average
PP/GF	Average	6,9	7,3	7,1	7,2	7,1
	St. Dev.	*0,46*	*0,17*	*0,41*	*0,35*	*0,2*
PP/BF	Average	6,2	6,5	6,5	6,6	6,4
	St. Dev.	*0,10*	*0,16*	*0,07*	*0,26*	*0,15*
PP+PP-g-MA/GF	Average	12,7	11,6	11,2	11,3	11,7
	St. Dev.	*1,12*	*0,14*	*0,35*	*0,18*	*0,7*
PP+PP-g-MA/BF	Average	11,6	11,7	11,7	11,8	11,7
	St. Dev.	*0,23*	*0,45*	*0,70*	*0,29*	*0,10*

Table 4. Notched Charpy impact resistance for samples injected at different conditions.

On the other hand, as can be seen in Figure 3 injection moulding parameters have little influence on the impact properties of the composites.

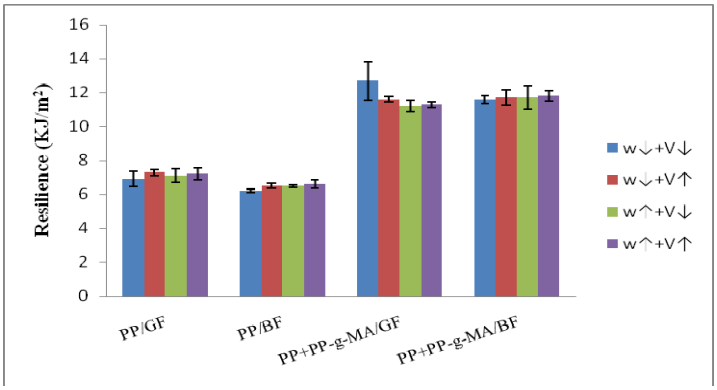

Figure 3. Notched Charpy impact resistance for samples injected at different conditions.

3.2. Fiber length distribution

Fibre lengths followed a normal distribution for all strand and samples, as can be observed in Figure 4. The mean lengths and the standard deviation were calculated for each analysed material and no significant differences were appreciated between strand (pre-pelletization) samples and injection moulded specimens.

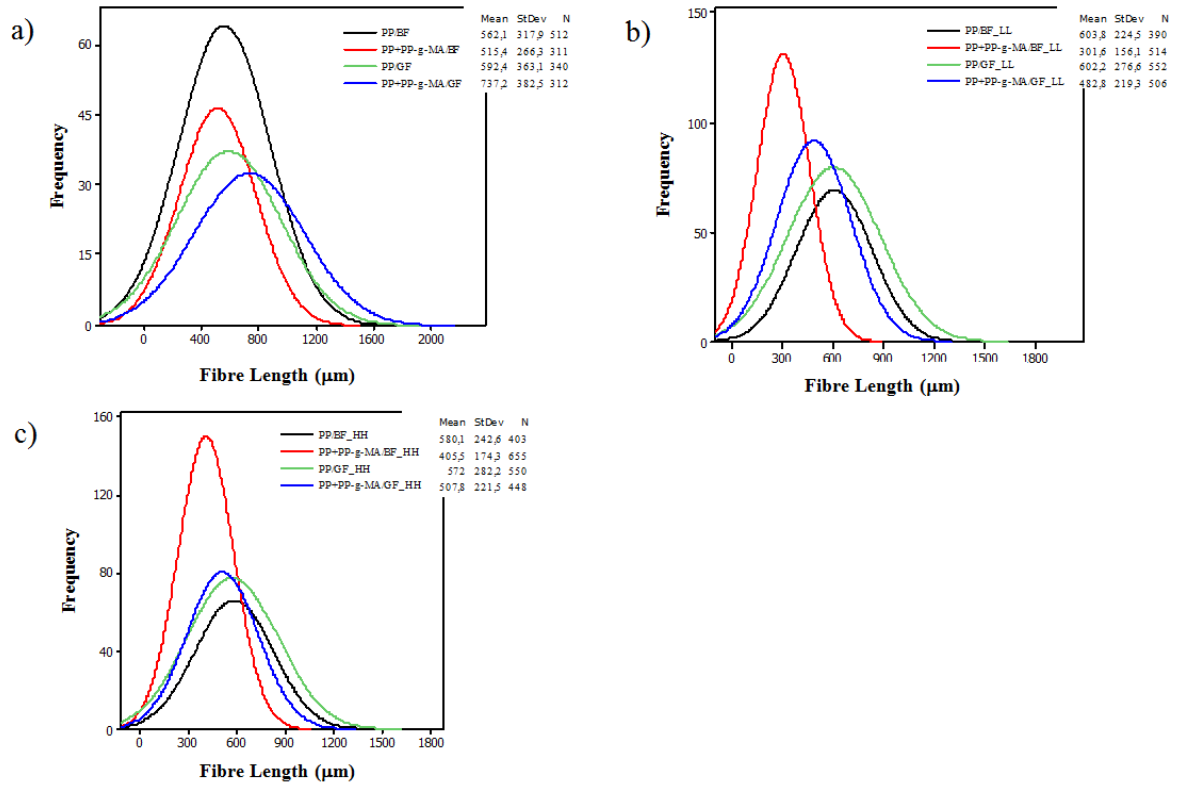

Figure 4. Normal distribution of fibre length of a) composite strands, b) specimens injected at high plasticisation and injection rate and c) specimens injected at low plasticisation and injection rate.

Fibre lengths of pellets were not measured because they were supposed to be similar to those of pre-pelletized strand and injection moulded specimens' lengths.

3.3 Fibre-matrix interphase. Effect of the coupling agent

Micrographs in Figure 5 show evidences of the huge influence of the maleic anhydride on the matrix-fibre interphase. Samples without coupling agent (1a, 1b) show the poor adhesion between the polypropylene and the basalt fibre. A clear gap between the matrix and the fibres is visible. On the contrary, samples with PP-g-MA (2a, 2b) show that the polymer surrounding the fibres holds them during tensile test, giving rise to an improvement on the mechanical properties of the composites.

The behaviour of the maleic anhydride is similar for BF and GF. The addition of MA greatly improves the matrix/fibre interphase (4a, 4b). However, according to the micrographs and mechanical properties, the coupling effect of maleic anhydride seems to be more important for BF containing composites than for the GF reinforced ones. The former seem to have a better interphase while the latter show less polymer surrounding the fibres, less adhesion, giving rise to a poorer improvement of mechanical properties.

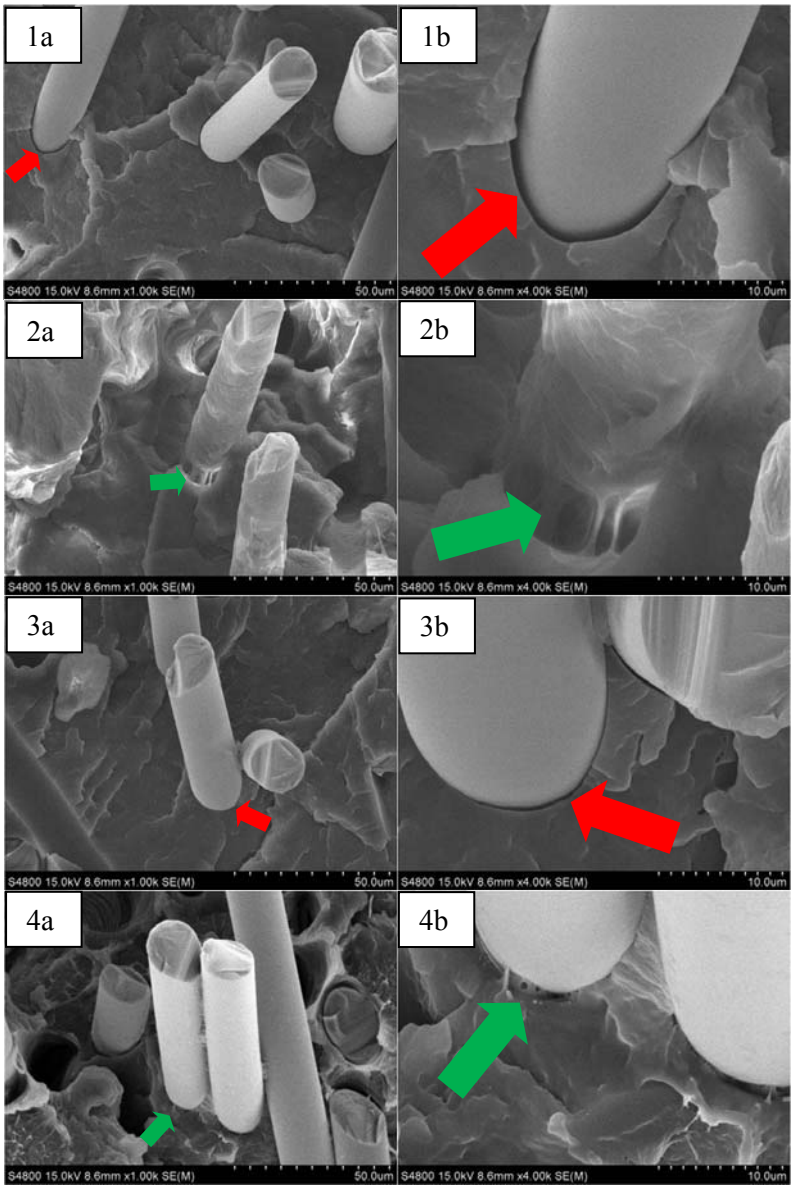

Figure 5. SEM micrographs of tensile tested specimens. 1a) PP/BF, low magnification; 1b) PP/BF, high magn.; 2a) PP+PP-g-MA/BF, low magn.; 2b) PP+PP-g-MA/BF, high magn.; 3a) PP/GF, low magn.; 3b) PP/GF, high magn.; 4a) PP+PP-g-MA/GF, low magn.; 1b) PP+PP-g-MA/GF, high magn.

4 Conclusions

Due to the poor interphase between the polypropylene matrix and the fibres (both glass and basalt), the reinforcement is not able to properly withstand the applied external stress. Thus, for those composites without coupling agent, the final tensile properties are mainly defined by the matrix's resistance, resulting in similar mechanical behaviour. However, taking into account the obtained impact resistance results, better values are observed for PP/GF composites than for PP/BF composites.

The incorporation of PP-g-MA has improved the interphase between the polypropylene matrix and fibres (both basalt and glass fibres) giving rise to an enhancement of the composite's tensile properties (tensile strength and deformation) and impact resistance. The

coupling agent works in such an efficient way that the tensile strength and impact resistance are improved up to around 80% (see tables 2 and 3). PP+PP-g-MA/BF composites show better overall mechanical behaviour than the other composites, including PP+PP-g-MA/GF samples.

On the other hand, the neat PP's elastic modulus is around 1,6 GPa and the incorporation of 30% in weight of GF or BF increases it up to 7 GPa. The addition of MA grafted PP slightly enhances this value, but the influence is almost negligible because Young's modulus is mainly improved just by the fibre content.

Surprisingly, there was not found a direct relationship between injection parameters and mechanical properties in the studied injection speed and plasticisation range. This was probably because the main fibre length that was obtained after extrusion was too short to still keep breaking when injected. Thus, different injection parameter values (high and low plasticisation and/or injection speeds) do not seem to have any influence on the final fibre length. Therefore it is not possible to match the obtained fibre lengths with different mechanical properties such as tensile strength. If longer fibres had been obtained after extrusion, injection parameters might have had influence on the breaking and so on the mechanical properties.

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