REGENERATION OF THERMALLY GLASS FIBRE FOR COST EFFECTIVE COMPOSITE RECYCLING: THE EFFECT OF FIBRE REGENERATION AND MATRIX MODIFICATION

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ABSTRACT

The recycling of glass fibre composites has become an important issue because of social-ecological, financial and legislative reasons. One main obstacle for the recycling of glass fibre composites is the low value of the recycled fibres. The present study investigates methods to maximize the reinforcement potential of thermally recycled glass fibres. Glass fibres were exposed to thermal recycling temperatures and processed into injection moulded polypropylene composites. Microbond tests were performed to characterize the adhesion between the fibres and the polypropylene matrix. Similar to other studies, the composite performance dropped when the fibres were exposed to high temperatures. It was found that the regeneration of the interfacial shear strength using maleic anhydride grafted polypropylene improves the composites performance but a post treatment of the fibres with γ-aminopropyltriethoxysilane was more effective. It was concluded that the application of γ-aminopropyltriethoxysilane might have improved the strength of the fibres.

1 INTRODUCTION

The glass fibre composites industry has experienced a rapid growth over the last decades and the global annual production of glass fibre composites is expected to exceed 10 millions tons this year. More than 50% of the glass fibre composites are thermoset based which are not intrinsically recyclable like thermoplastic based composites. The recycling of glass fibre composites was less interesting than the recycling of carbon fibre composites because of the higher value of carbon fibres [1]. Currently, glass fibre composites are often disposed as landfill but the recycling of glass fibre composites has become more desirable because of social-ecological, financial and legislative reasons. Societies and governments have become aware of the negative effects of landfill. Consequently, landfill is banned in some countries (e.g. Germany). In other countries, the governments raise taxes on landfill [1,2]. The European Union has released the 'End of Life Vehicles' directive which forces car manufacturers to increase the recyclability of cars to 85% by 2015 [3].

Thermal recycling processes like the fluidized bed [4] process can provide relatively clean and long fibres without the use of hazardous chemicals. One major disadvantage is that glass fibres lose their strength and sizing during thermal recycling processes [5]. Thus thermally recycled glass cannot compete with pristine glass fibres. Yang et al. [6] recently demonstrated that reinforcement potential of thermally recycled glass fibres could be improved by a post treatment with hydrogen fluoride. The present work looks at other ways that do not involve the use of hazardous chemicals like hydrogen fluoride to maximize the reinforcement potential of thermally recycled glass fibres.

2 EXPERIMENTAL

2.1 Fibre treatement

DS2200-13P chopped glass fibres with Polypropylene compatible sizing were provided by 3B Fibreglass company. The nominal diameter of the fibres is 13μm and the nominal length 4mm. The

fibres were thermally conditioned before composite processing in a ‘Carbolite CWF 12/13’ furnace at 500°C for 25min in air. After thermal conditioning the fibres were allowed to cool down in air. The thermally conditioned fibres were immersed for 15min in 1vol% γ-amino- propyltriethoxysilane (APS) solution. The solution was prepared by mixing the APS with deionized water 24h before use. After chemical treatment the fibres were dried at 110°C for 4h.

2.2 Composite processing

The composites were processed via extrusion compounding and injection moulding. The glass fibres were extrusion compounded with ‘SABIC® PP 579 S’ Polypropylene (PP) in a ‘Betol BC25’ single screw extruder. ‘Polybond 3200’ maleic anhydride-grafted polypropylene (MaPP) was added to the composites to promote the adhesion between glass fibres and matrix. The extruded material was drawn through a water bath and cut into pellets using a rotary cutter. The pellets were fed into an ‘Arburg 170-90/200’ injection moulding machine to produce dog-bone shaped tensile test specimens according to ASTM 638. All samples were stored for three weeks at room temperature conditions before mechanical testing. The nominal fibre content of all composites was 30wt%.

2.3 Mechanical testing

Tensile tests were performed using an ‘Instron 5969’ testing machine. All tests were performed with a head displacement rate of 1mm/min. A video extensometer was used to measure the strain. At least 5 samples of each batch were tested.

2.4 Fibre length measurement

Untested tensile test specimens were ashed in a programmable ‘Carbolite CWF 12/13’ furnace to separate the glass fibres from the PP matrix. Then the fibres were dispersed in deionized water and poured into petri dishes. The petri dishes were scanned by a high resolution scanner and the scanned image was analysed using an ‘IDM FASEP’ fibre length analysis system. At least 10000 fibres were analysed for each sample.

3 RESULTS AND DISCUSSION

3.1 Tensile strength and micromechanical properties

Figure 1 shows the tensile strength of the investigated composites. Figure 1 shows no difference between composites based on as received fibres with 1wt% added MaPP and 8% added MaPP. In contrast, the tensile strength of the composites based on thermally preconditioned fibres improved significantly when 8 wt% MaPP were added to the composites. Both composites based on thermally conditioned fibres did not approach the tensile strength of the composites based on as received fibres. When the fibres were post treated with APS after thermal conditioning the tensile strength of the composites almost reached the level of the composites based on as received fibres.

Preliminary work of this study showed that the tensile strength of composites based on as received fibres improves due to the addition of MaPP but reaches a plateau when more than 0.5wt% MaPP are added to the composite. This increase of the tensile strength can be attributed to an improvement of the adhesion between fibre and matrix. The as received fibres were fully sized with a PP compatible sizing. Thus a relatively low amount of MaPP was needed to optimize the tensile strength and the addition 8 wt% MaPP did not lead to an increase of the tensile strength. Similar observation on sized fibres were reported by Bowland [7] when he investigated the tensile properties of long glass fibre PP composites. The thermally preconditioned fibres lost most of their sizing during the heat treatment and larger amounts of MaPP were required to optimize adhesion between fibre and matrix. Rijsdijk et al. [8] investigated the effect of MaPP on unsized fibres and observed that 10wt% MaPP were required to optimize the tensile properties of long glass fibre PP composites. The lower tensile strength of the composites based on thermally conditioned fibres might be explained with the loss of the PP compatible sizing and a reduction of the fibre strength. Microbond tests and micromechanical analysis were used to investigate how the MaPP and APS influence the properties of the composites.

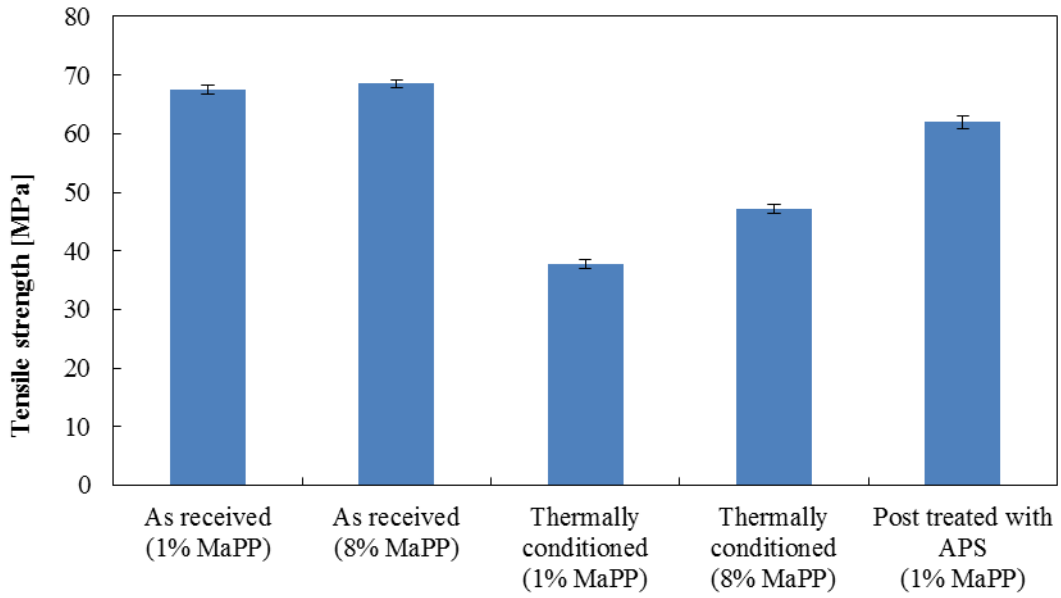

Figure 1. Tensile strength of the studied composites

Figure 2 shows the values for the IFSS that were obtained from the microbond tests and the fibre stress at composite failure that was determined from the tensile test data using the Kelly-Tyson model. As expected the IFSS between as received fibres and PP with 1wt% added MaPP content is higher than the IFSS between heat treated fibres and PP with the same amount of added MaPP. When the MaPP content was increased to 8wt% the measured IFSS between the heat treated fibres and the PP approached almost the value of as received fibres. Similar to the addition of 8wt% MaPP, the post treatment of the heat treated fibres with APS also increased the measured value for the IFSS. The

microbond test results suggest that the IFSS between heat treated fibres and PP with 8wt% added MaPP might have been similar to the IFSS between post treated fibres and PP with 1wt% added MaPP.

Fu et al. [9] demonstrated that the amount of residual polymer on fibres cannot always be used as a measure for the interfacial adhesion. However in the present study, the fracture surfaces in Figure 3 support the results of the microbond tests. The as received fibres were covered with residual PP while the heat treated fibres were relatively clean when only 1wt% MaPP was added to the PP. Residual PP was also found on fibres when 8wt% MaPP were added to the composites and when the fibres were post treated with APS.

Although the microbond test results suggest similar levels of adhesion the composites based on post treated fibres were significantly stronger than the composites based on heat treated fibres with a high MaPP content. The fibre stress at composite failure plotted in Figure 2 shows that the post treated fibres carried similar loads like the as received fibres at composites failure. The addition of MaPP did only slightly improve the fibre stress at composite failure.

It is currently not clear why the post treatment of the fibres with APS improved the composite properties more than the addition of MaPP. One possible explanation might be that the APS heals flaws of the thermally conditioned fibres. It was previously reported that APS can heal flaws in glass fibres [10,11]. The data of the present study might support this theory. However, these studies investigated as received fibres that were not exposed to thermal recycling temperatures. A more recent study by Yang and Thomason [12] showed that the positive effect of APS on the strength of glass fibres might be explained from the surface protection point of view. Kennerly et al. [13] recoated glass fibres after thermal conditioning in air at 625°C for 20min with γ-methacryloxypropy- ltrimethoxy silane. They did not observe an improvement of the fibres strength. Sáez-Rodríguez et al. [14] thermally conditioned glass fibres similar to the present study for 25min at 500°C in air. They did not observe an increase of the fibre strength when the thermally conditioned fibres were sized with APS. However, they performed single fibre tensile tests on fibres with 20mm gauge length. The residual length of the fibres after composite processing was measured to be less than 500µm. Thus the results might not be comparable due to gauge length effects and APS might have a positive impact on the strength of very short fibres.

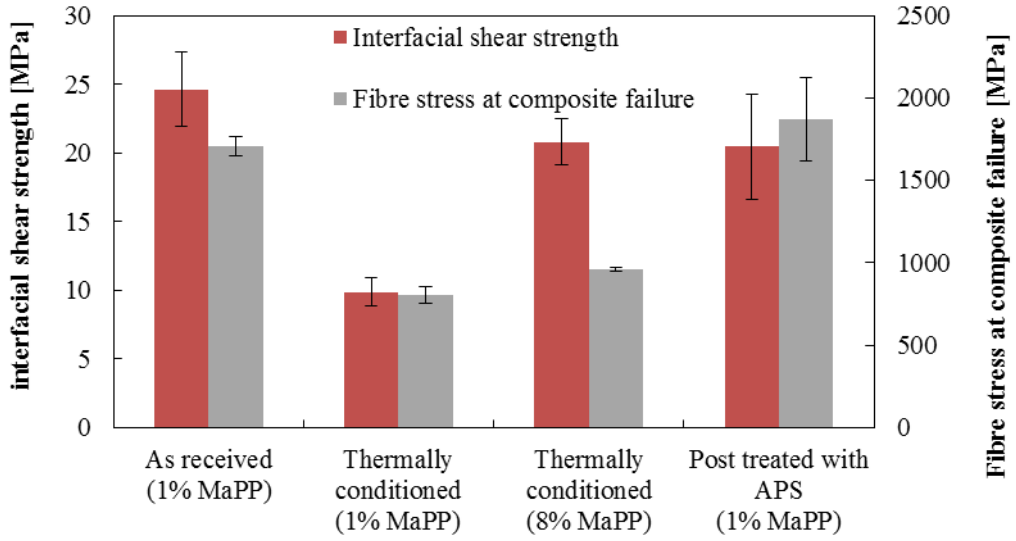

Figure 2. Interfacial shear strength and fibre stress at composite failure obtained from microbond tests and micromechanical analysis

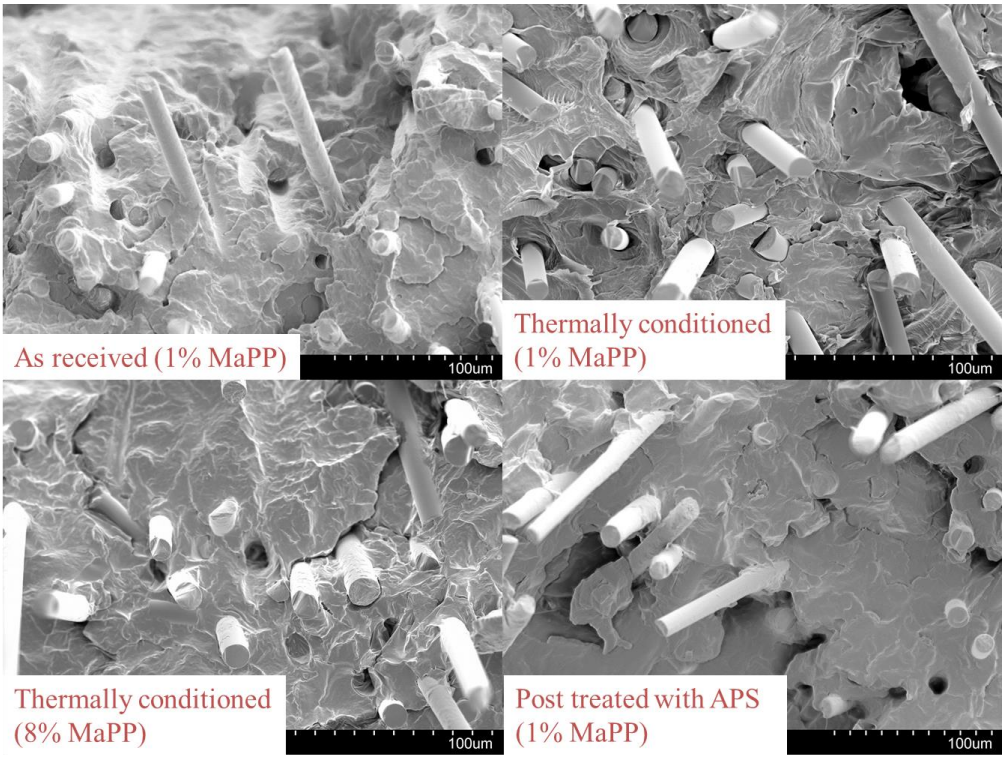

Figure 3. SEM micrographs of the tensile tested composites

3.2 Failure strain

The failure strain in Figure 4 behaved partly similar to the tensile strength in Figure 1. In contrast to the tensile strength, the failure strain of did not improve when 8wt% MaPP were added to the composites based on thermally conditioned fibres but the post treatment of the thermally conditioned fibres regenerated the failure strain of the composites completely.

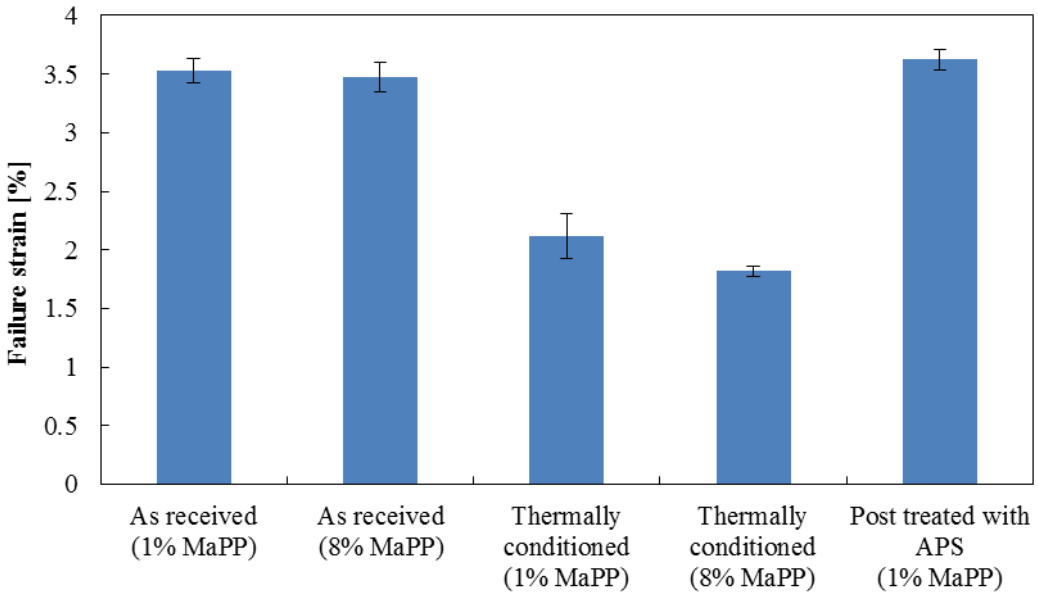

Figure 4. Failure strain of studied composites

3.3 Young's modulus

The values for the Young's modulus of the investigated composites and residual length of the glass fibres after composite processing are listed in Table 1. A reduction of the Young's modulus was observed when the glass fibres were exposed to thermal recycling temperatures before composite processing. The Young's modulus of discontinuous glass fibre polypropylene composites is known to be dependent on the residual fibre length, the modulus of the glass fibres, the fibre orientation and fibre content [15–19]. All investigated composites were processed under the same conditions and the same fibre content was used. Several researchers reported that the modulus of glass fibres increases [20,21] or does not change [22,23] due to exposure to thermal recycling temperatures. It is therefore likely that the reduction of the Young's modulus of the composites was caused by a reduction of the residual fibre length. It was previously shown that the length degradation of glass fibres during hot melt processing is influenced by the tensile strength of the fibres [24,25]. Thus the reduction of the residual fibre length could partly be explained with a reduction of the glass fibre strength. In addition, the residual fibre length might have been influenced by the arrangement of the fibres. The chopped fibres were received as bundles but broke up easily into single fibres after thermal conditioning. Single fibres were previously observed to be more susceptible to length degradation than fibres bundles during hot melt processing [5]. The arrangement of the fibres and the relatively thin layer of sizing might also explain why the post treatment with APS did not lead to an increase of the residual fibre length.

Fibre reinforcement and added MaPP content	Young's modulus [GPa]	Residual fibre length [µm]
As received (1% MaPP)	6.21±0.26	344
Thermally conditioned (1% MaPP)	5.93±0.37	315
Thermally conditioned (8% MaPP)	5.50±0.13	315
Post treated with APS (1%MaPP)	5.40±0.36	302

Table 1. Young's modulus of investigated composites

4 CONCLUSION

The present study demonstrates that thermally recycled glass fibres need a post treatment to act as effective reinforcement in polypropylene (PP) composites. The adhesion between fibres and polypropylene can be regenerated by the addition of maleic anhydride grafted polypropylene (MaPP). The optimization of the interfacial adhesion also led to an improvement of the composite performance. However, the post treatment of the fibres with γ-amino- propyltriethoxysilane (APS) was more effective in improving the composite performance. The composites based on the post treated fibres almost reached the performance of the composites based on as received fibres. This might indicate that APS can improve the strength of glass fibres after exposure to thermal recycling temperatures if the gauge length is sufficiently short. Thus a post treatment with APS might be sufficient to regenerate the reinforcement potential of thermally recycled glass fibres for the application in injection moulded PP composites.

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