

REGENERATION OF THERMALLY RECYCLED GLASS FIBRE FOR COST-EFFECTIVE COMPOSITE RECYCLING: PERFORMANCE OF COMPOSITES BASED ON PP AND RECOVERED GLASS FIBRE

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Keywords: glass, composites, recycling, regeneration

Abstract

Due to economic and technical reasons, no recycling process for glass fibre composites has been commercialized on a large scale. Thermal recycling processes are promising in terms of potential for commercialization but the reinforcement potential of thermally recycled fibres is too low for the application in composites. In the present study, glass fibres were exposed to elevated temperatures prior to composite processing to imitate a thermal recycling process. The exposure of the fibres to elevated temperatures prior to composite processing caused a significant reduction of the mechanical properties of the composites. The heat treated fibres were regenerated with a post treatment. The regeneration of the glass fibres recovered the mechanical properties of the composites almost completely. Thus, this study shows that composites based on thermally recycled glass fibres have the potential to compete with composites based on 'new' glass fibres.

1. Introduction

The development of economically viable and environmentally sustainable recycling processes for glass fibre composites has become highly important. 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 fluidised bed method are interesting because they do not involve the use of hazardous substances like chemical recycling processes and they can produce cleaner fibres than mechanical recycling processes. A major disadvantage of thermal recycling processes is the reduction of the glass fibre strength due to high temperatures [1,2,4]. A successful recovery of the strength of thermally recycled glass fibres could enable such recycled fibres to compete with, and replace, pristine materials in many large volume composite applications. The reuse of these materials could result in a huge reduction in the environmental impact of the glass-fibre composites supply industry.

Composites based on thermally recycled glass fibres have been produced before [5] but they showed a poor mechanical performance and were therefore economically not interesting. The present study shows that composites based on thermally recycled glass fibres can compete with composites based on pristine glass fibres if the fibres are regenerated.

2. Experimental

2.1. Fibre treatment

The present study is based on chopped ‘DS 2200-13P’ glass fibres that were provided by 3B fiberglass company. The glass fibres were heat treated prior to composite processing to imitate a thermal recycling process. The heat treatment of the glass fibres was performed in a ‘Carbolite CWF 12/13’ furnace. The furnace was preheated for 1h to allow the temperature to stabilize prior to inserting the fibres. The fibres were placed in an aluminum tray and heat treated in the furnace at 500°C. After 25min the fibres were allowed to cool down under room temperature conditions.

A ReCoVer treatment was applied to the heat treated glass fibres before the composite processing to enhance the mechanical properties of the composites. Discussion of the ReCoVer treatment will be presented in a parallel ECCM paper [6]. In this paper, the heat treated glass fibres that were regenerated with the ReCoVer treatment are called regenerated glass fibres. The glass fibres that were incorporated into the composites as received from the manufacturer are called untreated fibres.

2.2 Composite Processing

A ‘Betol BC25’ extruder was used to compound ‘SABIC® PP 579 S’ Polypropylene (PP) pellets with the glass fibres. 1% ‘Polybond 3200’ maleic anhydride-grafted polypropylene by PP weight was added to the composites to improve the adhesion between glass fibres and PP matrix. The extruded material was drawn through a water bath and cut into pellets using a rotary cutter.

An ‘Arburg 170-90/200’ injection moulding machine was used to produce dog-bone shaped tensile test specimens according to ASTM 638.

The processing temperatures were set between 170°C and 230°C. All samples were conditioned at room temperature for three weeks before mechanical testing.

2.3 Mechanical testing

An ‘Instron 5969’ testing machine was used to perform the tensile tests. The strain was recorded with a video extensometer. A constant head displacement rate of 1mm/min was used to test the composites. The PP was tested with a head displacement rate of 1mm/min up to 3% strain. Then the head displacement rate was increased to 5mm/min.

Unnotched Charpy impact test specimens according to ISO 179-1 were cut from injection moulded tensile bars. A ‘Tinius Olsen Impact 503’ impact tester with a 25J hammer was used to perform the impact tests.

2.4 Fibre length measurement

The glass fibres were extracted from the injection moulded tensile test specimens by ashing in a 'Carbolite CWF 12/13' furnace. Temperatures up to 500°C were used to burn off the PP completely.

An 'IDM FASEP' fibre length analysis system was used to measure the length of the extracted fibres.

2.5 Interfacial shear strength and fibre stress at failure

Bowyer and Bader [7] developed an algorithm to obtain the interfacial shear strength (IFSS) from tensile test data of injection moulded glass fibre polypropylene (GF/PP) composites. The algorithm is based on the Kelly-Tyson model. Thomason [8,9] improved and extended the algorithm to calculate the fibre stress at failure. The same algorithm was used in the present study to calculate the IFSS and fibre stress at failure.

The extended Kelly-Tyson model describes the tensile stress in composites with equation 1

$$\sigma_c = \eta * (X + Y) + Z \quad (1)$$

where η describes the fibre orientation, X is the contribution of the subcritical fibres and Y is the contribution of the supercritical fibres. Z is the contribution of the matrix.

The contribution of all subcritical fibres X with the volume subfraction V_i , fibre length l_i , fibre radius r and interfacial shear stress τ can be described with equation 2.

$$X = \sum \frac{\tau * l_i * V_i}{2 * r} \quad (2)$$

The contribution of the supercritical fibres Y with the volume subfraction V_j , length l_j , fibre radius r and can be described with

$$Y = \sum E_f * \varepsilon_c * \left(1 - \frac{E_f * \varepsilon_c * r}{2 * l_j * \tau}\right) * V_j \quad (3)$$

where E_f is the Young's modulus of the fibre and τ is the interfacial shear strength. ε_c stands for the strain of the composite.

Based on the equations above the following algorithm was used to calculate the IFSS and the fibre stress at failure in the present study.

(1) First the ratio R was determined with equation 4. σ_1 and σ_2 are tensile stresses in the composite at two chosen strains ε_1 and ε_2 ($\varepsilon_2 = 2 * \varepsilon_1$). Z_1 and Z_2 are the matrix contributions which were calculated with equation 5. Equation 5 describes the stress in the matrix as a function of the strain and was determined experimentally.

$$R = \frac{\sigma_1 - Z_1}{\sigma_2 - Z_2} \quad (4)$$

$$Z = 18.7 * \varepsilon^3 - 4.7 * \varepsilon^2 + 0.4 * \varepsilon \quad (5)$$

(2) In the second step, a value for the IFSS was chosen and the critical fibre length at the strains ε_1 and ε_2 was calculated with equation 6.

$$l_\varepsilon = \frac{E_f * r_f * \varepsilon_c}{\tau} \quad (6)$$

(3) Then the contributions X_1 , X_2 of the subcritical fibres and the contributions of the supercritical fibres Y_1 , Y_2 at the chosen strains ε_1 and ε_2 were calculated with equation 2 and equation 3. The assumed IFSS and critical fibre length were used as input parameter. The length of glass fibres was measured and the value for the stiffness of the fibres was taken from literature [10]. A fibre diameter $13\mu\text{m}$ ($r=6.5\mu\text{m}$) was quoted by the manufacturer.

(4) In the next step, the theoretical value of the ratio of the fibre contribution R' at the chosen strains ε_1 and ε_2 was calculated and compared with the measured value R .

$$R' = \frac{X_1 + Y_1}{X_2 + Y_2} \quad (7)$$

(5) The value of the IFSS was adjusted until the theoretical value R' matched the experimental value R . The adjusted value for the IFSS was regarded as the true value. After the determination of the IFSS, the orientation factor could be determined by rearranging equation 1.

(6) The fibre stress at failure was calculated using the IFSS from (5) as input parameter. A value for the fibre stress at failure σ_F was chosen and used to calculate the critical fibre length at composite failure with equation 8.

$$l_c = \frac{\sigma_F * r}{\tau} \quad (8)$$

(7) The calculated value for the critical fibre length at composite failure and the calculated value for the IFSS were used in equation 1-4 to calculate the composite stress at failure. The value for the fibre stress at failure was adjusted until the calculated value for the composite stress at failure matched the measured tensile strength of the composite.

3. Results and discussion

3.1. Tensile properties

Figure 1 below compares the tensile strength of unreinforced polypropylene (PP) with glass fibre polypropylene (GF/PP) composites. The GF/PP composites were based on untreated glass fibres, heat treated glass fibres and regenerated glass fibres. In the present study, the tensile test data is based on at least 5 tensile tests. The error bars represent a 95% confidence limit.

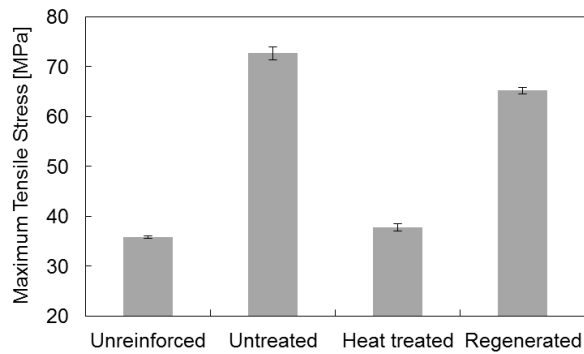


Figure 1. Tensile strength of PP and GF/PP composites based on pristine, heat treated and recovered glass fibres

As reported previously [11], the tensile strength of the GF/PP composites dropped almost to the strength of unreinforced PP when the glass fibres were preconditioned at 500°C. The regeneration of the glass fibres improved the tensile strength of the composites significantly. Thus more than 70% of the strength loss was recovered due to the regeneration of the glass fibres.

Figure 2 shows the failure strain of composites based on untreated glass fibres, heat treated glass fibres and regenerated glass fibres.

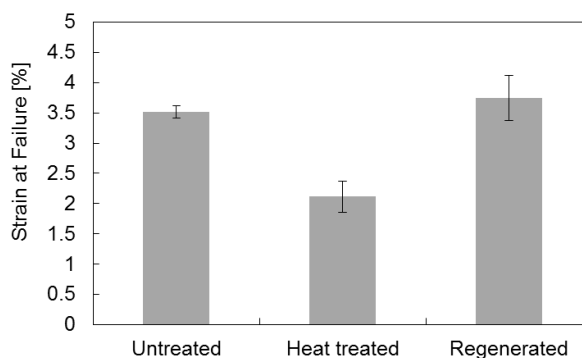


Figure 2. Failure strain of PP and GF/PP composites based on pristine, heat treated and recovered glass fibres

As reported previously [11], the failure strain of composites dropped when the glass fibres were heat treated before the composite processing. The regeneration of the heat treated glass fibres led to a complete recovery of the failure strain.

3.2 Impact properties

Figure 3 below compares the unnotched charpy impact strength of unreinforced PP, with GF/PP composites based on untreated glass fibres, heat treated glass fibres and regenerated glass fibres. Each data point is based on 10 specimens. The error bars represent a 95% confidence limit.

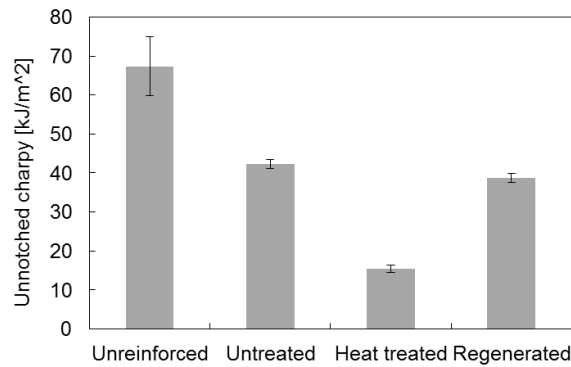


Figure 3. Unnotched Charpy impact strength of PP and GF/PP composites based on pristine, heat treated and recovered glass fibres

The data in Figure 3 shows that the unnotched Charpy impact strength of the composites was significantly reduced due to the heat treatment of the glass fibres prior to composite processing. More than 80% of the impact strength was recovered when the heat treated fibres were regenerated.

3.3 Interfacial shear strength and fibre stress at failure

Heat treated glass fibres are more susceptible to length degradation during extrusion compounding and injection moulding which leads to a shorter residual fibre length in the composites. This phenomenon can be explained with a degradation of the fibre strength and the arrangement of fibres when they are fed into the extruder [11]. In the present study, the length weighted average of the residual length of the untreated glass fibres was measured to be 479µm. The weighted average length of the heat treated fibres and the regenerated fibres was measured to be 442µm and 446µm respectively.

In the present study, the reduction of the tensile strength, failure strain and unnotched Charpy impact strength cannot be explained with a reduction of the fibre length because the residual fibre length is below the critical fibre length. These properties are dominated by the interfacial shear strength (IFSS) [11].

Figure 4 and Figure 5 below show the IFSS and fibre stress at failure that were calculated from the tensile test data above. The thermal preconditioning caused a significant drop of the IFSS. The IFSS was almost completely recovered when a ReCoVer treatment was applied to the heat treated glass fibres.

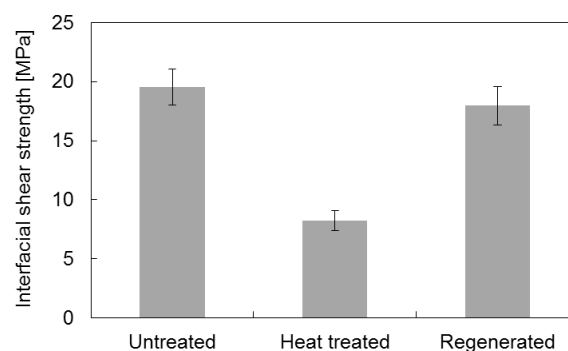


Figure 4. IFSS of GF/PP composites based on pristine, heat treated and recovered glass fibres

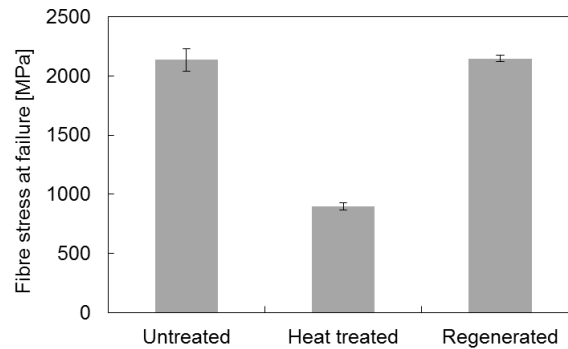


Figure 5. Fibre stress at failure of GF/PP composites based on pristine, heat treated and recovered glass fibres

Similar to the IFSS, the fibre stress at failure dropped when the glass fibres were heat treated. The fibre stress at failure was recovered when the ReCoVer treatment was applied to the glass fibres.

Several studies showed that the heat treatment of glass fibres leads to a significant reduction of the glass fibre tensile strength [12,13,14]. The fibre stress at failure cannot be regarded as fibre strength because the composite performance is also influenced by the IFSS. However, the values in Figure 5 indicate that the regenerated fibres in the composites can carry the same load as untreated fibres.

4. Conclusion

In the present study, glass fibres were heat treated before the composite processing to imitate a thermal recycling process. The composite performance was reduced to the level of the unreinforced matrix due to the fibre heat treatment. The regeneration of the glass fibres recovered the composite performance almost completely.

It can be concluded that in injection moulded polypropylene composites, thermally recycled glass fibres have the potential to replace new or untreated glass fibres. The performance of the composites based on recycled glass fibres can reach the performance of composites based on untreated fibres if the fibres are regenerated after the thermal recycling process.

Acknowledgements

This work is part of the EPSRC grant ‘Towards Affordable, Closed-Loop Recyclable Future Low Carbon Vehicle Structures’ (TARF-LCV). The financial support by the EPSRC is gratefully acknowledged. The authors would also like to thank the Advanced Materials Research Laboratory (AMRL) for the use of the mechanical testing machines and the SEM. The help of 3B fibreglass company and Saudi Basic Industries Corporation (SABIC) for the supply of the materials is also acknowledged.

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