

DEVELOPMENT OF BULK MOULDING COMPOUNDS AND EXTRUSION COMPOUNDING PROCESSES USING RECYCLED GLASS FIBRES

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Abstract

Project PRoGrESS aims to deliver a pilot-scale fluidised bed recycling facility to recover the glass fibre reinforcement from end-of-life composite material which can then be reprocessed, moulded, and reused in relevant industrial applications. In this paper, model and genuine fibre recyclates were used to produce bulk moulding compounds and injection moulding test materials. It was found that BMC products with mechanical properties acceptable for commercial implementation could be achieved with almost total replacement of virgin glass fibres with heat-treated glass fibres, though increased fibre content was necessary. Application of a coupling agent had little significant effect on BMC mechanical properties and the introduction of an extended mixing period significantly reduced tensile, flexural, and impact properties. Compounding of recycled glass fibres with polypropylene showed that the fluffy fibres tended to form entangled mats unsuitable for extrusion. Compounding at levels of reinforcement loading above 10 wt.% was not possible due to poor material flow in the hopper. It is concluded that in addition to optimising recycling technologies and fibre post-treatment formulations, suitable manufacturing or fibre preparation techniques must be developed to support the introduction of recycled materials into conventional thermoplastic composite manufacturing processes.

1. Introduction

To achieve global carbon neutrality, adoption of renewable energy sources such as wind power can be expected to rise significantly in the coming decades. As demand for wind power increases, so does the significant amount of end-of-life composite blade waste generated during decommissioning [1]. Consequently, developing a cost-effective and environmentally conscious method of recycling large composite structures continues to be an essential challenge to achieving a circular economy for the global composites industry and there remains a need for investment in waste facilities capable of handling these materials. Current disposal routes, such as landfill and incineration, do not recover materials for reuse and are already being phased out or subject to increasing legislation in several regions. At present, the level of technical maturity and industrial implementation of composite recycling varies between the available methods. Fluidised bed recycling involves thermal decomposition of the polymer matrix to liberate clean glass fibres into the gas stream which are subsequently separated and collected. The reclaimed fibres can then be reprocessed into new composite materials, reducing landfill burden and offsetting emissions associated with virgin material production through replacement.

The fluidised bed is a promising route for scalable composite recycling and has reduced global warming potential compared to conventional disposal routes such as landfill. Yang *et al.* report that fluidised bed recycling may achieve relatively low recycling costs with less energy consumption by 2050 [2]. Conversely, energy intensive recycling methods such as solvolysis and pyrolysis are more expensive than landfill and established end-of-life routes such as cement kiln co-processing may not be financially viable. Composite wind turbine blade feedstocks have good scope to be used in fluidised bed recycling methods due to their high glass fibre content enabling high levels of material recovery and relatively low resin content minimising GHG (greenhouse gas) emissions during thermal decomposition [3]. Electricity generation from fluidised bed recycling methods can further offset emissions, though the impact of this factor is reduced as electricity generation becomes increasingly decarbonised. The use of recycled glass fibres in the production of new composites can reduce the environmental impact and costs associated with the production of virgin glass fibres and scales with the amount of virgin material that is effectively displaced. However, it is essential that acceptable composite performance and material processability is achieved to support wider usage of these materials in the composite value chain.

Project PRoGrESS aims to deliver a pilot-scale fluidised bed recycling facility and encourage reuse of recycled glass fibres in suitable products throughout the composites supply chain [4]. Both bulk moulding compounds and injection moulded thermoplastics are promising routes for reuse of recycled glass fibres in new composite applications compared to long fibre laminates reliant on fibre lengths and tensile strengths that are often not achievable with scaled composite recycling processes [5,6]. In this paper, heat-treated model glass fibres were used to produce bulk moulding compounds (BMC) at varying ratios of virgin glass fibre replacement. Recycled glass fibres reclaimed from end-of-life turbine blade material using a fluidised bed were extrusion compounded with injection moulding grade polypropylene and a maleic anhydride functionalized homopolypropylene. The effect of fibre post-treatment with respective matrix-compatible silane coupling agents and processing conditions on composite mechanical properties are also reported.

2. Experimental Methods and Materials

2.1. Materials

Heat-treated virgin glass fibres exposed to elevated temperatures representative of a thermo-oxidative recycling process such as a fluidised bed were used in the production of bulk moulding compounds [3]. Approximately 330 g of 6 mm Owens Corning Advantex P316 chopped glass fibres were placed into an aluminium foil tray then transferred to a pre-heated furnace. Heat treatment was performed for 25 min at 500°C using a Carbolite CWF1200 electric muffle furnace before being removed and allowed to cool to room temperature. To improve compatibility with the polyester matrix, heat-treated glass fibres were resized with γ -methacryloxypropyltrimethoxysilane (MPTMS). A dilute acetic acid solution was used to adjust the pH of deionised water to between pH 5 and 5.5 in order to promote hydrolysis of the silane in water. A 1 vol.% MPTMS silane solution was prepared and hydrolysed for 12 h. The silane solution was then poured into the aluminium trays of heat-treated glass fibres and soaked for 15 min. The silane solution was then drained from the trays and fibres were dried for 12 h at 110°C.

Recycled glass fibres recovered from end-of-life wind turbine blade material using a laboratory scale fluidised bed reactor operating between 500–550°C were used to produce injection moulding pellets [7]. Wind blade material was received part shredded and was further downsized using a Zerma GSL slow speed granulator with a 5 mm screen before fluidised bed processing. Recycled glass fibres were resized with γ -aminopropyltriethoxysilane (APS) to improve adhesion with the polypropylene matrix. Approximately 300 g of recycled glass fibres were completely immersed in 4 L of a 1 vol.% silane solution for 15 min, removed from the solution and dried for 12 h at 110°C. The thermoplastic polymer materials used for the extrusion compounding were SABIC PP 579S and Exxelor PO1020.

2.2. Bulk Moulding Compounds

Bulk moulding compounds were produced using a laboratory scale twin sigma blade mixer according to an undisclosed commercial formulation comprised of polyester resin, calcium carbonate filler,

20 wt.% 6 mm chopped glass fibre reinforcement, and other additives. Filler, polymer, and additives were mixed for 20 min at high speed before adding glass fibres and mixing at slow speed for a further 3 min. The virgin glass fibre content in BMC formulations was replaced by 25–100% heat-treated model recycled glass fibres. Untreated model recycled glass fibres and fibres post-treated with a polyester compatible MPTMS coupling agent were investigated. The effect of BMC processing variables was also investigated by mixing at high speed for an additional 50 min. Test plaques (250 mm x 250 mm x 4 mm) were compression moulded according to ISO 1268-8 and mechanical test specimens were machined from the plaques using a waterjet cutter. Three plaques were moulded for each test series and specimens were cut at 0° and 90° orientation with respect to the longitudinal direction of the plaque.

2.3. Thermoplastic Compound Extrusion

A PRISM TSE 16 TC 16mm co-rotating twin-screw extruder was used to compound SABIC PP 579S polypropylene with 10–30 wt.% silanised recycled glass fibres and 7 wt.% Exxelor PO1020 maleic anhydride functionalised homopolypropylene. Compound extruded material was drawn through a water bath and subsequently cut into pellets using a granulator with a 5 mm screen.

2.4. Composite Testing

Tensile testing was performed according to ISO 527 using type 1B specimens. A constant crosshead speed of 1 mm/min was used and strain was recorded with a clip on extensometer. Flexural testing was performed according to ISO 14125 and a testing speed of 50 mm/min was used to maintain experimental parity with reference materials. Tensile and flexural tests were carried out using a Testometric X500 universal testing machine with a 50 kN load cell. Unnotched Charpy flatwise impact testing was performed according to ISO 179. Impact tests were carried out using a Tinius Olsen IT503 impact tester with a 25 J hammer. Ten specimens were tested for both flexural and impact measurements and four specimens were tested for tensile measurements.

3. Results and Discussion

3.1. Effect of silane treatment on BMC mechanical properties

Figure 1 shows the tensile, flexural, and impact properties of BMC composites with 0–100% substitution of the 6 mm virgin chopped glass fibres with heat-treated glass fibres subjected to a thermal history comparable to the fluidised bed recycling process. Both flexural and impact strength of BMC decreased linearly as more virgin glass was replaced by heat-treated fibres. This behaviour can largely be attributed to thermally induced loss of as much as 80% of the initial glass fibre tensile strength and removal of the original fibre size formulation [8,9]. BMC tensile strength was not significantly affected by replacement with heat-treated fibres, though baseline values are relatively low to begin with. Recent analysis by Yang *et al.* indicates that fluidised bed recycling processes may achieve net zero primary energy demand with a substitution rate as low as 40% due to the GHG intensity of the production of virgin glass fibres [2]. It is worth noting that, even at full replacement with unsized heat-treated fibres, flexural strength of approximately 60 MPa and impact strength of around 12 kJ/m² were achieved. These mechanical properties are acceptable for the production of high-volume non-structural BMC products such as electric meter boxes that typically use around 15% glass fibre in their formulations. Accordingly, the mechanical testing results for BMC formulations including heat-treated fibres indicate good scope for commercial application in infrastructural products. Depending on BMC product requirements it may be possible to achieve a balance between the amount of virgin material that is replaced by glass fibre recyclates reduced mechanical properties may be compensated by increasing the amount of glass fibre used in the BMC formulation.

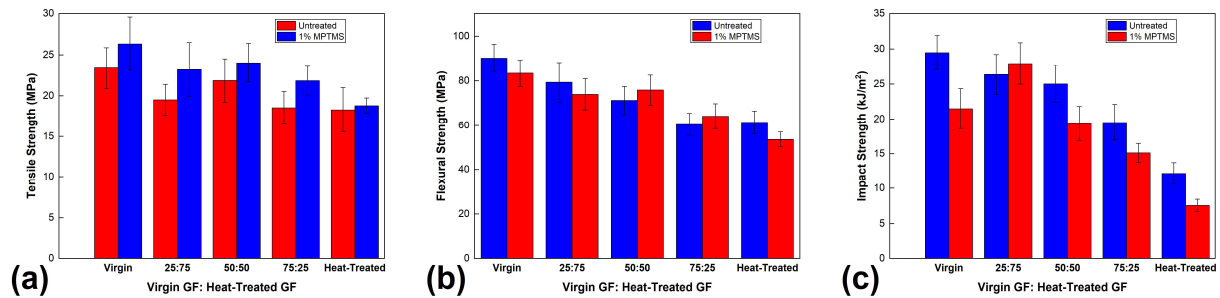


Figure 1. (a) Tensile, (b) flexural, and (c) impact properties of BMC with 0–100% replacement with unsized and silanised heat-treated glass fibres

Work by Yang *et al.* has indicated that application of silane coupling agent alone was able to recover 30–35% of the flexural strength loss of thermally degraded discontinuous glass fibre/epoxy composites [9]. In this study, we found that application of polyester compatible silane coupling agent to the thermally degraded glass fibres had little significant effect on tensile, flexural, or impact properties. The results show good agreement with the findings of Kennerley *et al.* in that a silane coating did not contribute to recovering the mechanical properties of dough moulded composites [6].

It is well established that silane coupling agents are the primary contributors to adhesion in glass fibre sizings. It is also known that re-application of a silane to thermally degraded fibres can restore much of the loss in interfacial adhesion. Figure 2 shows BMC plaques made with virgin and silane coated heat-treated glass fibres. It is visible that silane treated glass fibres dispersed poorly in the moulding compound compared to the virgin fibres and distinct bundle structures were apparent. Accordingly, it may be possible that any positive benefits of increased adhesion are offset by poor dispersion of the silane-treated fibres. It may also be possible that the concentration of the silane solution was too high. Future work will investigate the influence of silane concentration and the addition of dispersing aids. Fibre rinsing stages after the silane treatment may also be advisable. However, the inclusion of additional steps complicates implementation of such processes at scale.

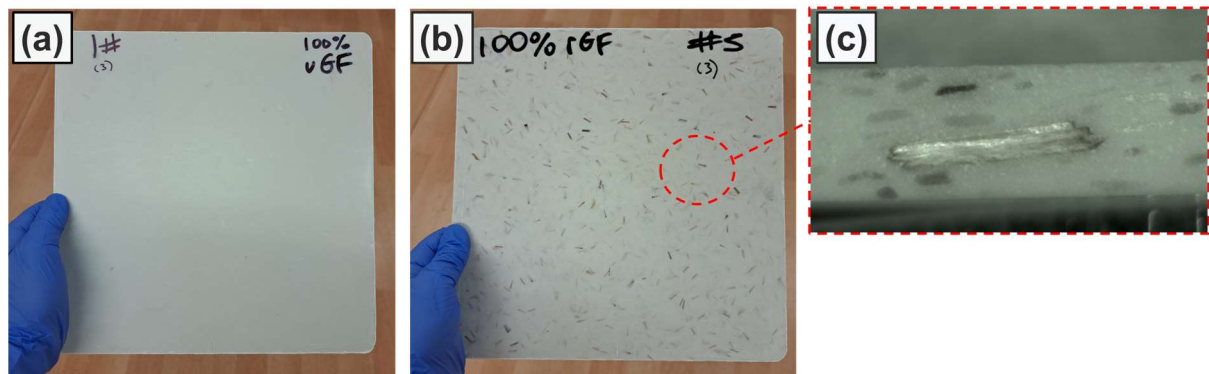


Figure 2: BMC plaque made with (a) virgin glass fibres (b) silane coated heat-treated glass fibres (c) optical micrograph of poorly dispersed fibre bundle

3.2. Effect of mixing time on BMC mechanical properties

An extended BMC mixing time was investigated to improve heat-treated glass fibre dispersion. Furthermore, studies by Faudree *et al.* have suggested that reducing fibre lengths to the sub-millimetre range through an extended mixing time can improve tensile and impact properties of BMC products [10,11]. This effect has been attributed to a combination of factors relevant to three-phase (polymer, fibre, filler) composites including: increased tolerance to microcracks, homogeneous distribution of increased thermal compressive stress sites, and the number of spaces between fibres per mm³ being increased by an order of magnitude. In this study, filler, polymer, and additives were combined into a

paste and mixed for 20 min at high speed before adding glass fibres and mixing at slow speed for a further 3 min followed by an additional 50 min mixing at high speed. It was observed that introduction of an extended mixing period caused the moulding compound made with heat-treated fibres to form distinct spherical structures during mixing (Figure 3a). This structure was reflected in the compression moulded test plaques and was not observed with virgin fibres (Figure 3b).

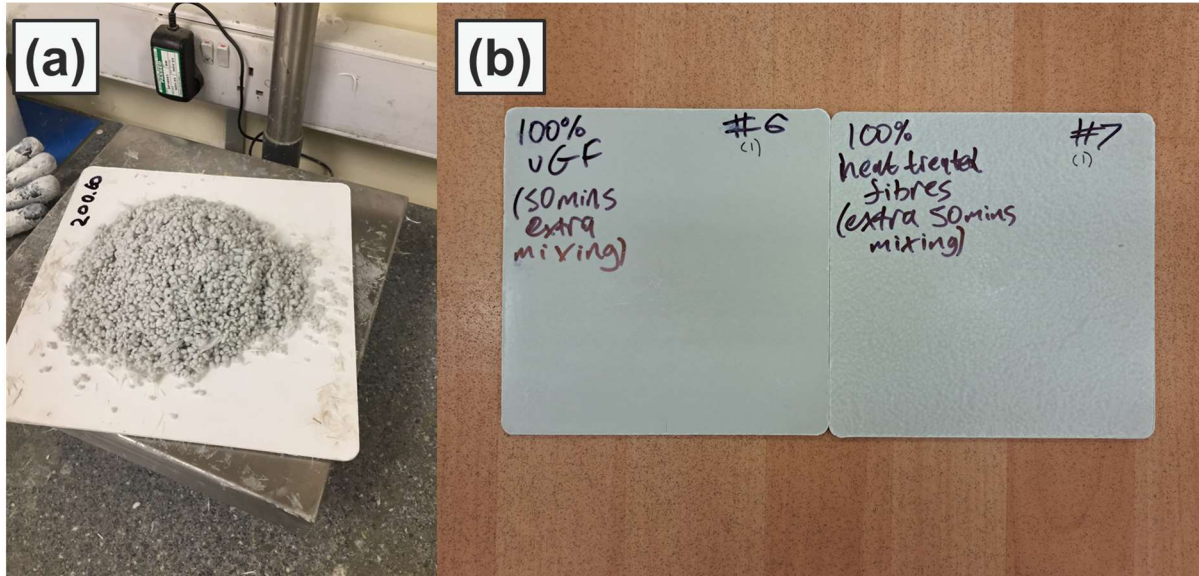


Figure 3: (a) Distinct spherical structures formed during extended mixing of BMC with heat-treated fibres and (b) comparison of virgin and silanised heat-treated glass fibre test plaques

Mechanical testing results for 100% virgin and 100% heat-treated glass fibres with MPTMS silane treatment following an extended paste mixing time are shown in Figure 4.

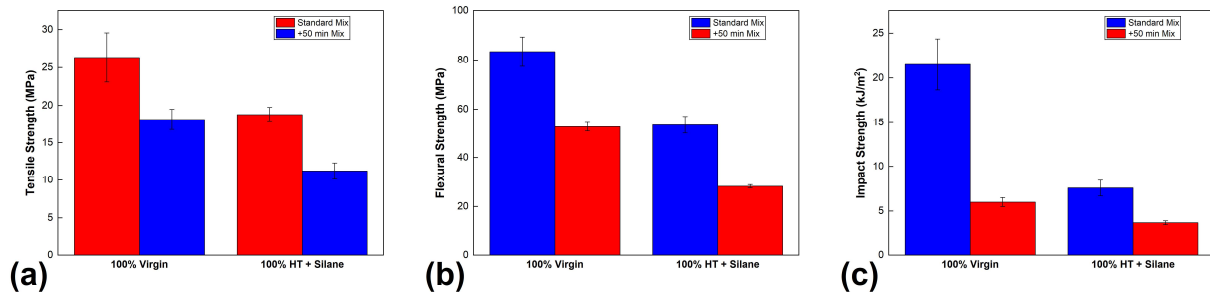


Figure 4: a) Tensile, b) flexural, and c) impact properties of BMC with 50 min extended mixing time

We report findings contrary to those in [10,11], in that the introduction of an extended mixing period reduced tensile, flexural, and impact properties for both virgin and heat-treated test plaques. It may be possible that the heat-treated fibres were poorly dispersed in the agglomerated mixture and stress concentrations in the specimens were induced by the spherulitic moulding compound. Investigation of the optimisation of BMC batch processing parameters to achieve increased mechanical performance may still be a promising route for recycled glass fibres if conditions conducive to well dispersed short fibre lengths can be achieved. Further work will investigate the influence of recycled glass fibre length reduction prior to mixing.

3.1. Thermoplastic extrusion compounding of recycled glass fibres

Recycled glass fibres treated with an aminosilane coupling agent tended to form hardened agglomerated bundles following treatment and drying as shown in Figure 5a. Direct compounding of these fibres was not possible due to the large glass fibre clumps sticking in the hopper and feed chutes and failing to mix with the thermoplastic polymer. To address this, treated fibres were re-chopped to approximately 6 mm length bundles using composite shears before extrusion. After re-chopping, silanised recycled glass fibres were successfully extrusion compounded using a twin screw extruder at a target fibre loading of 10 wt.% and chopped into 5 mm pellets suitable for injection moulding applications. Thermogravimetric analysis indicated that the actual level of fibre reinforcement level achieved was approximately 8 wt.%. It was generally not possible to achieve fibre loading in the 15–30 wt.% range due to poor material flow in the hopper relative to virgin chopped glass products and agglomeration of the relatively low bulk density fibre bundles in the hopper.

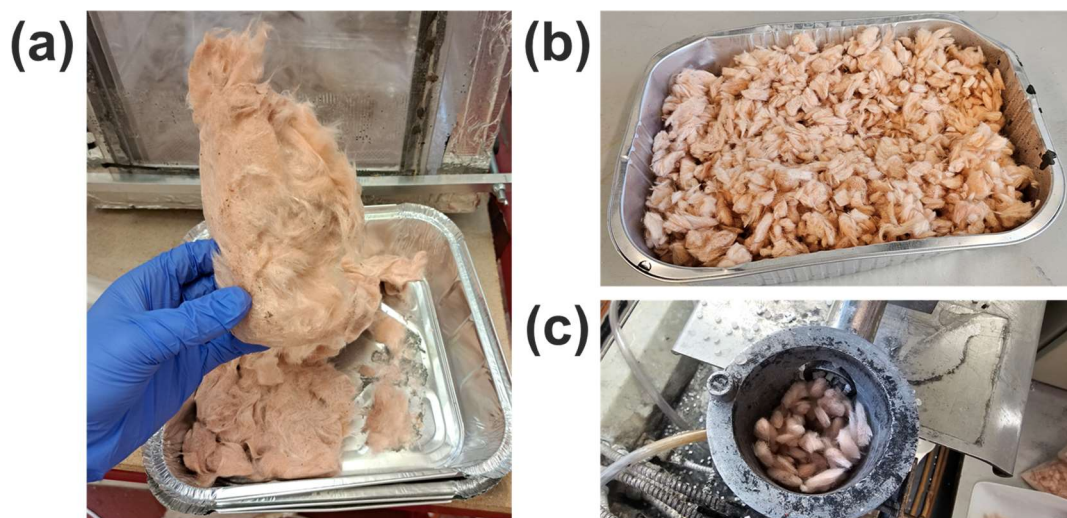


Figure 5: (a) Recycled glass fibres after aminosilane treatment and drying (b) after re-chopping to approximately 6 mm (c) agglomeration in extruder hopper

While optimisation of recycled glass fibre resizing processes remains a key parameter in delivering enhanced fibre and composite performance, a suitable process must be developed to support a “drop in” product for injection moulding applications that can be successfully compounded at higher levels of glass fibre reinforcement. This may be achieved through a combination of fibre treatments that enable enhanced reprocessing, optimisation of silane concentrations, and addition of lubricants or processing aids. Furthermore, the use of suitable technologies such as crammer feeders or side feeders may be necessary to facilitate usage of low bulk density recycled glass fibres at a scale suitable for commercial implementation.

4. Conclusions

In this paper, heat-treated model glass fibres were used to produce BMC at varying ratios of virgin glass fibre replacement and thermoplastic compounds were produced using recycled glass fibres. It was found that BMC products with mechanical properties acceptable for commercial implementation could be achieved with almost total replacement of virgin fibres. Depending on BMC product requirements it may be possible to achieve a balance between the amount of virgin material that is replaced by glass fibre recyclates and compensate reduced mechanical properties by increasing the amount of glass fibre used in the BMC formulation. Applying a polyester-compatible coupling agent had little significant effect on composite mechanical properties. This is possibly due to any positive benefits of increased adhesion being offset by silane-treated fibres being relatively poorly dispersed. The introduction of an extended mixing period caused the moulding compound to form distinct spherical structures and significantly reduced tensile, flexural, and impact properties were measured.

Recycled glass fibres treated with a silane solution were extrusion compounded with injection moulding grade polypropylene and a maleic anhydride functionalized homopolypropylene. Silane treated fibres tended to form hardened entangled mats unsuitable for compounding. The issue was mitigated by re-chopping treated fibres into bundles before extrusion to produce an injection moulding compound with approximately 10% fibre reinforcement. Extrusion compounding at higher levels of reinforcement loading was not possible due to poor material flow in the hopper relative to virgin chopped glass products. This was primarily caused by the low bulk density of recycled glass fibres and the tendency for treated fibres to agglomerate in the hopper. It is concluded that in addition to optimising recycling technologies and fibre post-treatment formulations, suitable fibre feeding process or fibre preparation techniques must be developed to support the introduction of recycled materials into conventional composite manufacturing processes.

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