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The effect of reprocessing on the mechanical properties of polypropylene reinforced with wood pulp, flax or glass fibre

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ABSTRACT

Composites of polypropylene, substitutable for a given application and reinforced with: Medium Density Fibreboard fibre (MDF) (40 wt%); flax (30 wt%); and glass fibre (20 wt%), were evaluated after 6 injection moulding and extrusion reprocessing cycles. Of the range of tensile, flexural and impact properties examined, MDF composites showed the best mean property retention after reprocessing (87%) compared to flax (72%) and glass (59%). After 1 reprocessing cycle the glass composite had higher tensile strength (56.2 MPa) compared to the MDF composite (44.4) but after 6 cycles the MDF was stronger (35.0 compared to 29.6 MPa for the glass composite). Property reductions were attributed to reduced fibre length. MDF fibres showed the lowest reduction in fibre length between 1 and 6 cycles (39%), compared to glass (51%) and flax (62%). Flax fibres showed greater increases in damage (cell wall dislocations) with reprocessing than was shown by MDF fibres.

1. Introduction

Increasing environmental awareness in the plastics industry has led to a need for waste management strategies. Political pressure has increased due to concerns about environmental sustainability and legislation is being put in place to divert material from landfill, if it is not considered as ultimate waste. For example, in the European Union the end of life vehicle directive regulates the disposal of vehicles. It has a target that 85% of the vehicle weight be either reused or recycled, and that a maximum of 5% is disposed in landfill [1]. To comply with the standards for environmental management systems (ISO 14001) and to manage costs, most plastics processors recycle the waste plastics generated during production (e.g.: runners, sprue, discarded parts and start/end of a run). A proportion of the ground wastes are incorporated with the original compound into the processing cycle; reducing costs and reducing the environmental impact of the process.

It is well known that processing composites damages fibres and reduces fibre length. Investigating commercial glass fibre reinforced polypropylene (PP) in extrusion and injection moulding processing Gupta et al. [2] showed that forces on the fibre, predominantly in the melting zone, break fibres into lengths of approximately 0.5 mm. It has been suggested that as the compounded polymer/fibre melts, the fibre is subjected to considerable shear and bending while still anchored in the melting polymer [3]. Breakage occurs when the bending load is higher than the failure load [4]. Further in the process when the polymer is molten, shear and elongation forces act on the fibre. Whether a fibre is broken depends on the magnitude of these forces and the fibre's length and its bending strength and stiffness [3,4]. In contrast to glass fibres which break once a critical stress is reached, natural fibres break after being submitted to repeated deformation cycles [5].

Guo et al. [6] compounded and injection moulded a range of natural fibres including wood pulps, bast fibres and wood flour. After just 1 injection moulding all fibre types were reduced to fibre lengths of approximately 0.25 mm. Multiple reprocessing cycles of fibre reinforced composites can bring about a further reduction in fibre length due to repeated exposure to high shear forces in extrusion and injection moulding. These reductions in fibre length are associated with reductions in mechanical properties in glass but relatively moderate changes in hemp and sisal reinforced composites [7]. Although the trend is usually a loss of mechanical properties with repeated reprocessing [8–11], reprocessing can also initially bring about an improvement in strength and stiffness attributed to the increased dispersal of the fibre within the matrix [8,12–16]. Extruded wood flour reinforced composites have shown reductions in flexural properties and little change in impact strength in polyvinyl chloride [17] but relatively stable tensile properties in PP [18]. During injection moulding, and especially

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with multiple cycles, the melt is submitted to high temperature and pressure, which are associated with high shear and elongational deformation rates. In plastics processing most fibre breakage occurs during screw processing as fibres are submitted to shear and bending forces [3,4].

With reprocessing, there is both mechanical and thermal degradation of the matrix and the natural fibre reinforcement. The thermo-mechanical effects of reprocessing are likely to degrade the polymer by random chain scissions in the polymer backbone resulting in molecular weight reduction and therefore loss in mechanical performance [19]. In addition, the process results in the thermal degradation of the natural fibre [20].

For effective and efficient composites manufacture and processing there are a range of considerations including: cost; runnability; and performance in-service. A range of different composite materials may be available and readily substitutable for a given application. The manufacturer will select a composite material for a part based on these considerations. In this work we compare the reprocessing of polypropylene composites made from fibre-rich pellets of Medium Density Fibreboard fibre (MDF), with comparable, and readily substitutable, flax and glass fibre composites. We have selected material with loadings (20% and 30% wt, for glass and flax respectively), expected to give comparable initial properties to composites made from 40% wood fibre. The purpose of this work is to investigate composites of comparable properties but different fibre reinforcements, and how reprocessing may affect their in-service performance. Both injection moulding and compound extrusion reprocessing were investigated. Only internal recycling, the reprocessing of waste material in the manufacturing process, is investigated in this study. It is not the purpose of this work to address the recycling of post-consumer products which are subject to more severe degradation caused by actual ageing or weathering before recycling.

2. Material and methods

2.1. Materials

A composite made of 40 wt% MDF wood fibre pellet reinforced PP was prepared by extrusion compounding with the following materials:

- Polypropylene matrix PP Seetec M1600 from LG Chem with 25 MFI (230 °C/2.16 kg).
- Maleic Anhydride Polypropylene (MAPP) G3015 from Eastman.
- MDF fibre pellets made of MDF wood fibre refined from *Pinus radiata* chips at the Scion mechanical pulping pilot plant following the process of Warnes et al. [21].

The flax composite was 30 wt% flax (*Linum usitatissimum*) fibre reinforced and compatibilised PP (NAFARU – Thuringian Institute for Textile and Plastic Research). This material was made via a textile process where endless twisted roast treated flax fibre bundles and continuous PP fibres were continuously stacked, orientated and stretched before the PP was melted via an IR heat process. After cooling, the bonded strands were cut into granules approximately 10 mm long. The material is compatibilised but the loading and formulation are not specified. The glass fibre composite was a commercially available, 20 wt% glass fibre, high impact resistance, reinforced PP (Lupol HI-2202 from LG Chem) for injection moulding applications [22]. This material is prepared by extrusion compounding and is compatibilised (the loading and formulation also not specified).

The term 'fibre' is used generically. Botanically speaking the MDF fibres are tracheids, the primary water conduction cells of the wood in conifers. The flax fibres are bast fibres, largely non-conducting cells of the phloem. Fibre volume fractions were estimated based on measured composite densities, PP density and fibre weight fractions. The estimated fibre volume fractions are 30%, 21% and 9% for the MDF, Flax and glass respectively.

2.2. Sample production

The MDF fibre pellets and 3 wt% MAPP were oven dried respectively at 100 and 60 °C overnight (~16 h) prior to extrusion. They were then combined with the polymer into a twin screw LABtech™ vented extruder type LTE 26–40 (26 mm co-rotating screws; l/d = 40). A mixture of PP/MAPP was fed "upstream" in the extruder by the main hopper. The MDF fibre pellets were fed "downstream" with the second feeder into the molten polymer. The compound went through a 4 strand die and was air cooled before being pelletized into 3 mm long pellets. The temperature of the melt was kept below 200 °C.

2.2.1. Injection moulding processing

The original compounded pellets of MDF, glass and flax fibre were injection moulded using a BOY 35 injection moulding machine into normalised tensile, flexural and impact test specimens. Twenty moulded specimens of each type were kept for testing and the remaining were granulated using a commercial plastics grinder into flakes which were then dried and injection moulded again. The above procedure was repeated 5 times. The 3 materials were injected at 180 °C with a mould temperature of 30 °C. The MDF and glass fibre composites were injected with a maximum injection pressure of 85 bars and a maximum injection speed of 80 mm/s. For the flax fibre composite, the injection pressure and speed was increased to 120 bars and 100 mm/s in order to get satisfactory samples because the molten compound seemed to have a higher viscosity.

2.2.2. Extrusion compounding processing

The original compounded pellets of MDF, glass and flax fibre were reprocessed by compound extrusion again on the Labtech twin screw extruder with similar conditions as above. The procedure was repeated 5 times. Cycle 1 is considered as representing the original properties of the compound without extrusion reprocessing. Injection moulded test specimens were produced with compounded pellets from cycles 1, 2, 4 and 6 using a BOY 35 M injection moulding machine as above. To investigate possible effects of reprocessing on the compatibiliser, fresh MAPP (2 wt%) was added prior to the 6th reprocessing cycle of a sub-sample of the MDF composites. As the compatibilisation details of the glass and flax composites were unknown, the appropriate addition of fresh compatibiliser was not possible.

2.3. Testing and evaluation

Tensile specimens were tested using an Instron 5566 machine. Tensile properties were measured according to ASTM D 638-03, type I test specimen with a crosshead speed of 5 mm/min. At least 5 specimens of each composite were tested to obtain the Young's modulus and the maximum tensile strength. Flexural properties were measured according to ASTM 790-03 with a crosshead speed of 1.3 mm/min. At least 5 specimens of every composite were tested to obtain the flexural modulus and the maximum flexural strength. The densities of the composites were calculated using a water displacement technique. Front notched Izod impact specimens were tested using a CEAST Resil impactor 6957 according to ASTM 256-06a standard. At least 5 specimens of each composite were tested to obtain the impact strength.