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Effect of coupling agents on reinforcing potential of recycled carbon fibre for polypropylene composite

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ABSTRACT

Polypropylene (PP) composites reinforced with recycled carbon fibre have been prepared through extrusion compounding and injection moulding. The reinforcing potential of the recycled fibre was increased by improving the interfacial adhesion between the fibre and PP matrix and this was done by the addition of maleic anhydride grafted polypropylene (MAPP) coupling agents. Three MAPP couplers with different molecular weights and maleic anhydride contents were considered. The effects on the mechanical properties of the composite were studied, and scanning electron microscopy (SEM) was used to study the fracture morphology of the tensile specimens. It was observed that with the addition of MAPP the interfacial adhesion was improved as fewer fibres were pulled-out and less debonding was seen. A microbond test was performed and a significant improvement in interfacial shear strength was measured. This resulted in composites with higher tensile and flexural strengths. The maximum strength was achieved from MAPP with the highest molecular weight. Increased modulus was also achieved with certain grades of MAPP. It was also found that the composite impact strength was improved significantly by MAPP, due to a higher compatibility between the fibre and matrix, which reduced crack initiation and propagation.

1. Introduction

The use of carbon fibre reinforced polymers is growing and diversifying. Their use is increasing in applications ranging from aerospace to sports equipment [1,2] and it was reported that the demand for carbon fibre in 2010 was 39,280 metric tonnes and will increase to 89,260 metric tonnes in 2015 [3]. Most of these fibres are used in thermosetting resins such as epoxy, phenolic and vinyl ester. Unlike their thermoplastic counterparts, the presence of a 3-dimensional cross-linked molecular structure in the thermosetting resin making recycling a difficult and challenging task that requires special processing techniques and technologies. Due to increasing environmental awareness, limited landfill capacity and enforcement of regulations for waste, a significant amount of effort and attention has been devoted globally to develop and exploit a variety of means for recycling these valuable materials [4,5].

It was reported that an energy intensity of 286 MJ/kg was required for making virgin carbon fibre, compared with only 33 MJ/kg for steel production [6]. This results in high manufacturing costs and so carbon fibre is limited in its use to areas where the high costs can be justified by a substantial gain in mechanical performance and weight saving. Recycled carbon fibres have the potential to be cheaper than virgin fibre and this could open up new markets and new opportunities in different industries. This has already been recognised and the development of a diverse range of applications has been proposed for recycled fibres in applications such as an electrically conductive materials for electromagnetic interference shielding [7], a thermally conductive fabric for heated clothing [8], a reinforcement for ceramic brake discs [9], electrode materials for fuel cell application [10] and a potential material for making activated fibre [11]. The focus of this study is to explore the feasibility of using a coupling agent to maximise the reinforcing potential of recycled fibre in a polypropylene composite. Polypropylene is produced in large quantities worldwide and is used extensively in the composite industries, such as for injection moulding, production of glass mat thermoplastics (GMTs) and long fibre thermoplastics (LFTs). In this work, attention is focused on injection moulding, as it accounts for a substantial part of the overall composites market [12]. A fluidised bed process [13] was used to recycle short carbon fibre from epoxy prepreg scrap. The fibre was then compounded and injection moulded with virgin polypropylene. To improve compatibility between the fibre and the non-polar matrix, maleic anhydride grafted polypropylene (MAPP) coupling agents of different acid numbers and molecular weights were added into the compounding process at different loadings. Tensile, flexural and impact tests on the prepared specimens were carried out. The adhesion between the fibre and matrix was studied via a microbond test together with an inspection on the tensile-fractured surfaces using SEM analysis.

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2. Experimental procedure

2.1. Materials

The polypropylene was supplied by Ineos Olefins and Polymers Europe, Lyndhurst, UK. It was 100-GA12, a general purpose grade homopolymer. According to manufacturer's data, it has a melt flow rate of 12 g/10 min at 230 °C/2.16 kg condition, tensile strength at yield of 34 MPa and flexural modulus of 1400 MPa. The number and weight average molecular weight is 37,733 and 283,000 respectively [14]. Three maleic anhydride grafted polypropylene (MAPP) coupling agents, supplied by Eastman, UK under the trade name of E43, G3015 and G3003, were used. The coupling agents were in a granular form and their physical properties, provided by the manufacturer, are given in Table 1. The content of the maleic anhydride functional group was measured via titration and it was expressed in term of acid number [15].

Scrap prepreg roll with a width of 0.6 m was supplied by Advanced Composites Group, Heanor, UK. It comprised an MTM28-2 epoxy resin reinforced with Toray T600SC-24000-60E unidirectional 24 k carbon fibre. To fully cure the prepreg, multiple sheets were cut to a dimension of approximately 0.6 m × 1.0 m and heated at 100 °C inside an oven for 5 h. The silicone paper was then removed and a guillotine was used to cut the prepreg sheets into ribbons of 12.0 ± 1.5 mm wide (measured along the fibre axis). The ribbons were torn manually along the fibre axis into rectangular pieces of length 30–60 mm. The rectangular pieces were then shredded using a Retsch SM2000 miller equipped with a 10 mm-by-10 mm square aperture sieve into irregular strips of width ranging from 0.5 to 3 mm and length 2 to 12 mm.

2.2. Fluidised bed recycling process

The shredded prepreg was fed into a fluidised bed process to recycle the carbon fibre. The prepreg was fed continuously at 2 g/min into a bubbling sand bed at 550 °C. The bed consisted of sand particles with an average diameter of 0.85 mm and it was fluidised with hot air with a fluidised velocity of 1 m/s. The epoxy matrix decomposed at this temperature and fibres were elutriated from the bed and separated from the gas stream in a cyclone. An afterburner with an operating temperature of 850 °C was used to fully oxidise any remaining volatile gases from the exhaust stream. The tensile property of the recycled fibre was determined according to BS ISO 11566:1996 standard. A gauge length of 6 mm was used. The tensile strength and modulus of the fibre were 3.03 GPa and 197.63 MPa respectively. The weight average fibre length was 8.65 mm. The average diameter of the fibre, d, obtained from a SEM analysis, was 7.56 ± 0.21 μm [7].

2.3. Manufacture of carbon fibre pellet

Carbon fibre recycled from the fluidised bed process was in a fluffy and discontinuous form. It was converted into a roll of random non-woven mat with an areal density of 100 g/m^2 using a papermaking type process by Technical Fibre Product Ltd., Cumbria, UK. The mat was slit into long narrow strips of 4 mm wide using an office paper shredder. The strips were then cut using a pneumatic chopping machine into pellets of 4 mm wide by 6 mm long.

Table 1
Physical properties of coupling agents.

Property	E43	G3015	G3003
Density (kg/m^3)	0.934	0.913	0.912
Acid number (mg KOH/g)	45	15	9
Number average molecular weight, M_n	3900	24,000	27,000
Weight average molecular weight, M_w	9100	47,000	52,000
Molecular weight distribution (MWD), M_w/M_n	2.33	1.96	1.93
Brookfield viscosity @190 °C, cP	300	18,000	60,000

2.4. Manufacture of composite material

Composite test specimens were fabricated via two processes: compounding and injection moulding. The compounding was undertaken using a co-rotating intermeshing twin-screw extruder supplied by Rondol, Stoke on Trent, UK. The screw diameter was 21 mm and its L/D ratio was 25:1. Two compounding processes were employed. In the first process, coupling agent and PP granules were tumble mixed before being fed into the main hopper of the extruder. A rotating stirrer was equipped inside the hopper for continuous mixing of the two materials. There were five temperature controlled zones along the screws and they were set to 210/210/210/200/190 °C, increasing from feeding to the exit zones. The mixture was extruded using a screw rotational speed of 80 rpm through a 3 mm diameter die, forming a continuous strand, which was then water-cooled, pelletised and dried. The residence time for the granules to travel from the feeding zone to the die outlet was 130 s. This pre-compounding process was repeated for preparing granules with different coupling agents at weight percentages of 2%, 5% and 8% respectively. In the second compounding process, the pre-compounded granules were reintroduced back to the extruder via the main hopper whilst carbon fibre pellets were fed into the extruder through a side feeder, forming a composite containing 30% by weight of fibre. The side feeder was located just after the melting zone of the barrel in order to minimise damage to the fibres through the combination of mechanical, thermal and chemical degradation [16,17]. The same 3 mm diameter die and temperature settings were used but the rotational speed was reduced to 50 rpm to minimise fibre damage. With this lower screw speed, the residence time within the screw barrel increased to 150 s. Two benchmarking materials were also included in this study. The first one comprised only the polypropylene and it was used as a control to demonstrate the reinforcing potential of the recycled fibre. The second benchmarking material contained recycled carbon fibre but without any coupling agent. All benchmarking materials were subjected to the same compounding processes so that a similar processing history was maintained.

A Battenfeld BSKM 170/46 DS-2000 injection moulding machine was used to fabricate a 2.4 mm thick multi-purpose dumb-bell shaped specimen, as specified by ISO 3167:1993. A melt temperature of 210/210/210/200 °C, increasing from the feeding to nozzle zones and a mould temperature of 50 °C were used. The hold pressure and back pressure were set to 12 MPa and 47.25 MPa respectively. At least 40 mouldings were fabricated for each formulation. To allow for temperature and material settling during the start of the moulding process, the first 15 mouldings were rejected. The dumb-bell shaped specimen was then machined and used for composite property characterisation as detailed in Section 2.5.

2.5. Composite characterisation

A microbond test, as illustrated in Fig. 1, was performed to measure the interfacial shear strength (IFSS), τ, between a microdroplet and a single fibre filament. The same twin-screw extruder was used to compound the coupling agent and PP granules under the same compounding conditions as specified in Section 2.4. However, instead of making chopped pallets, a continuous strand of diameter about 0.4 mm was extruded at the end of the second compounding process. The strand was cut into a length of around 6 mm, then slightly bent across its centre and transferred to a