PRODUCTION WASTE MANAGEMENT OF THERMOPLASTIC COMPOSITES USING COMPRESSION MOULDING

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ABSTRACT

This study focuses on the mechanical recycling technique applied to fibre reinforced thermoplastics. The material studied are production scrap and off-cuts of bi-directional woven glass reinforced polypropylene pre-consolidated laminates. The traditional mechanical recycling technique involves a shredding step, giving coarse grains, followed by a grinding step, giving a finer material which is afterwards used as reinforcement in thermoplastic injection processes. However, from those two steps, degradation issues related to the reinforcement effective length limiting the reused material to low-end applications are encountered. The concept under study in this research work consists in by-passing the grinding step and to keep only a shredding or cutting step so as to limit the degradation of the initial material potential and keep a substantial reinforcement length. The obtained grains/aggregates are then directly reprocessed in bulk form by compression moulding. This preliminary study showed that the modulus of the recycled composite demonstrated values close to the equivalent quasi-isotropic continuous fibre laminate of the same composition. However, a noteworthy decrease in the recycled composites' strengths were observed.

1 INTRODUCTION

Within the composite industry, the proportion of thermoplastic based composites is expected to grow in comparison to thermoset based composites. Indeed, the use of thermoplastic matrix offers some interesting intrinsic properties namely: long-term storage at room temperature, no transportation restriction, low toxicity and their potential of recyclability due to thermal reversibility. In this context, along with environmental legislation such as the European Union Waste Framework Directive, the waste management and recycling in this field has become a challenging topic of research. Several recycling techniques, namely pyrolysis, solvolysis and mechanical processes are being explored and optimised by the research community to increase their economical and environmental viability [1].

The traditional mechanical recycling involves a shredding step, giving coarse grains, followed by a grinding step, giving a finer material which is afterwards used as reinforcement in thermoplastic injection processes. This recycling chain offers the advantage of high production rate, complex shaped manufactured parts and low scrap material in the production line. However, from the shredding and grinding steps issues regarding the reinforcement effective length (1-3 mm) [2]. Furthermore, the injection process is generally limited to a reinforcement volume fraction below 40%, implying virgin material input. Low fibre volume fraction and short reinforcement length result in less rigidity, therefore limiting the recycled material to low-end applications. Recent research have shown that for this traditional mechanical recycling to be economically viable, the mechanical properties of the recycled material needs to be higher [1]. The scope of the present study is to assess the potential of an alternate mechanical recycling technique. This alternate technique, presented later on, is based on the advancement made in the manufacturing of thermoplastic composites this last decade.

In the past decade, extensive research has been conducted on compression moulding of chopped prepreg tapes in bulk form [3-17]. Interesting results have been obtained. Complex shaped parts have successfully been processed with high fibre volume fraction up to 61% with high mechanical properties. The chopped tapes have the parent material properties and since they are preimpregnated, no additional material input is needed in the production line. These so-formed materials exhibit mechanical properties which are dependent on the constitutive parameters as well as the grain or aggregate arrangement generated during manufacturing [11, 18, 19]. This manufacturing method gives the possibility to have long fibre lengths and high volume fraction and is a good compromise between high performance continuous fibre composite which are hardly formable in complex shapes and low performance, low fibre volume short fibre composites processed in complex shapes.

Our goal in this study is to apply this concept of bulk forming to process scrap of thermoplastic composites. That is, instead of having chopped tapes as material input, aggregates of laminates of thermoplastic composites are used. The obtained aggregates are then directly reprocessed in bulk form by compression moulding without additional material. The tensile properties of the recycled panels using this concept are assessed in order to evaluate the potential of this recycling technique.

2. MATERIALS AND METHODS

2.1. Materials

The initial form of the material in the production line is a glass/polypropylene (PP) laminate with a fibre volume fraction of 47 % (Figure 1a). Within the preconsolidated laminate, the reinforcement is in the form of a balanced 2x2 twill weave fabric. The tensile modulus and strength in the principal directions is 20 GPa and 395 MPa respectively. The laminates are 1 mm thick.

The production scrap are cut-offs of the initial laminate (Figure 1b). For the purpose of this study, rectangular aggregates of sides 25 mm x 5 mm were cut from the production scrap (Figure 1c).

a) b) c)

Figure 1. a) Initial laminate, b) Production scrap, c) aggregates.

2.2. Panel manufacturing

The composite panels were manufactured in a 300 mm by 300 mm shear-edge compression mould (Figure 2a). Aggregate-reinforced composites panels of 3 mm in thickness (Figure 2b), were manufactured according to two different charge ratio, 100 % (Figure 3a) and 75 % (Figure 3b). For the comparison of the tensile properties, quasi-isotropic (QI) $[0°/45°/-45°/90°]_{2s}$ continuous fibre reinforced panels were manufactured as well.

The mould is charged in material and closed till a contact pressure is measured. The mould is then subjected to a heat ramp of 5°C/min till a temperature of 200°C is reached. A temperature plateau at 200°C is then maintained for 15mins in order to have an isothermal condition in the mould cavity. After 10 mins on the temperature plateau, a consolidation pressure is applied (3 bars for QI panels and

30 bars for aggregate-reinforced panels) till the end of the thermo-compression process. At the end of the temperature plateau, a cooling ramp of 5°C/min is applied to the mould till the temperature of 100°C is reached and the panel unmolded.

a) b)

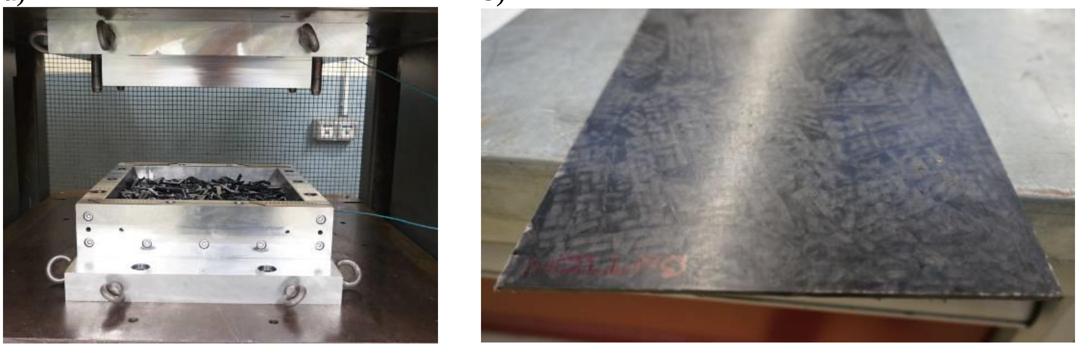

Figure 2. a) Compression mould, b) aggregate-reinforced composite panel.

a) b)

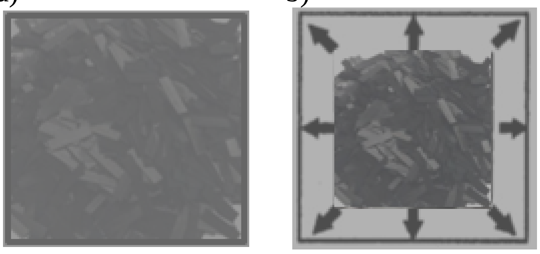

Figure 3. Charge ratio a) 100%, b) 75%.

2.3. Mechanical testing

Tensile test specimens of dimension 250 mm by 25 mm, according to ISO 527-4 [20], were cut from the aggregate-reinforced composites panels for testing. A speckle pattern was created on each test specimen in order to follow the displacement field using the DIC technique (Figure 4). The specimens were loaded at a rate of 1mm/min. The average strain field in the gauge section was used to plot the stress-strain curve.

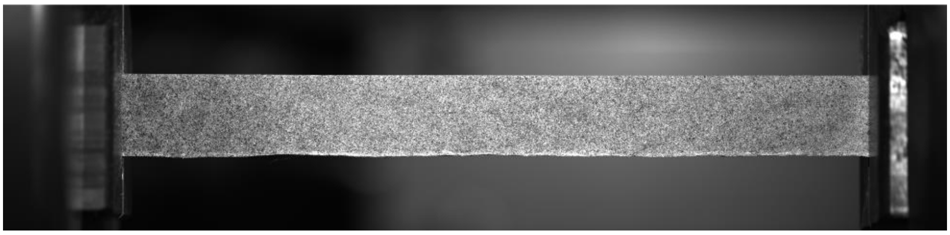

Figure 4. Speckle on tensile test specimen.

3. RESULTS AND DISCUSSION

3.1. Tensile behaviour

A typical stress-strain curve obtained on specimen from aggregate-reinforced recycled panels is shown on Figure 5. The stress rises linearly as a function of the axial strain and then deviates to a non-linear behaviour as a result of damage propagation in the specimen till a maximum stress value is reached.

The monitoring of the strain field at different stages of loading (Figure 6), reveals many regions where larger strain values are localised, even at low loads (Figure 6a). The high strain regions can be associated to resin-rich regions or regions of low reinforcement presence due to the discontinuities of the aggregates in the specimen. It can also be noted that the high strain regions arise mainly from the edges of the specimen. This has been observed by Selezneva et al. [14] on chopped tape reinforced composites as well. With the increase of the tensile load, high strain regions continue to increase throughout the specimen (Figure 6b), till it causes the final failure of the specimen.

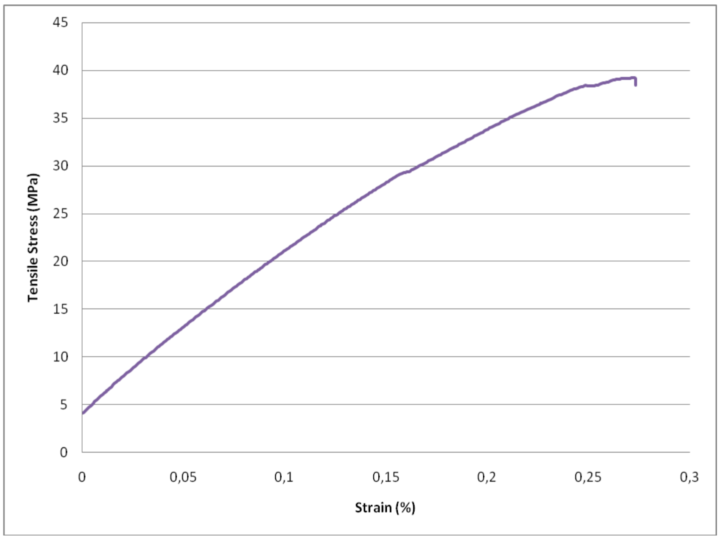

Figure 5. Tensile stress-strain curve of recycled specimen.

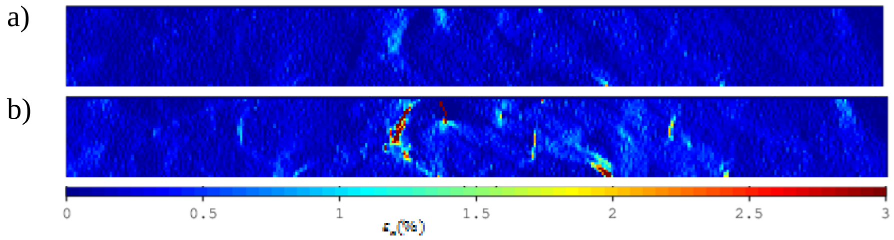

Figure 6. Strain field in gauge section of tensile specimen a) at 20 MPa, b) at 36 MPa.

The fracture faces of a tensile specimen is shown on Figure 7. For this specimen, the crack initiated from a resin-rich regions in the upper part of the fracture face. The crack then propagated throughout the specimen while avoiding the surrounding aggregates through longitudinal splitting and delamination in the matrix. This crack path can be attributed to stress concentration at the vicinity of the aggregates. As a result, no fibre failures were observed on the fracture faces indicating that the failure is exclusively dominated by the matrix.

Figure 7. Fracture faces of recycled tensile specimen.

3.2. Tensile Properties

The average tensile properties measured on the different panels are shown on the bar graphs on Figure 8. The values obtained on the quasi-isotropic continuous laminate, the aggregate-reinforced panel with a 100% charge ratio and the aggregate-reinforced panel with 75% charge ratio are respectively denominated as QI, RA100 and RA75 on the graphs. The average values are normalised to the tensile properties of the QI laminate taken as reference. The error bars represent the maximum and the minimum values measured in each batch.

Regarding the average tensile modulus measured on each panel (Figure 8a), it can be seen that the tensile modulus of the recycled aggregate-reinforced composites are comparable to that of the continuous QI panel with a decrease of -18 % and -8 % respectively for the RA100 panel and the RA75 panel. The RA100 panel and the RA75 panel only differ in charge ratio. Test samples from the RA75 showed a higher modulus compared to the specimens tested from the RA100 panel. It can be inferred that the macroscopic flow that takes place during the moulding of the RA75 panel induces a mesoscopic flow as mentioned in [11, 18] that results in more entanglement between the aggregates within that panel compared to the RA100 panel. The higher degree of entanglement reduces resin-rich regions and also lowers the heterogeneity by providing continuity between contiguous domains of aggregates within the panel [21]. This continuity between the contiguous domains of aggregates could explain the higher modulus measured on the RA75 panel and the lower scattering of the tensile modulus within the batch. A good proportion of the tensile modulus can therefore be recovered after recycling using this method.

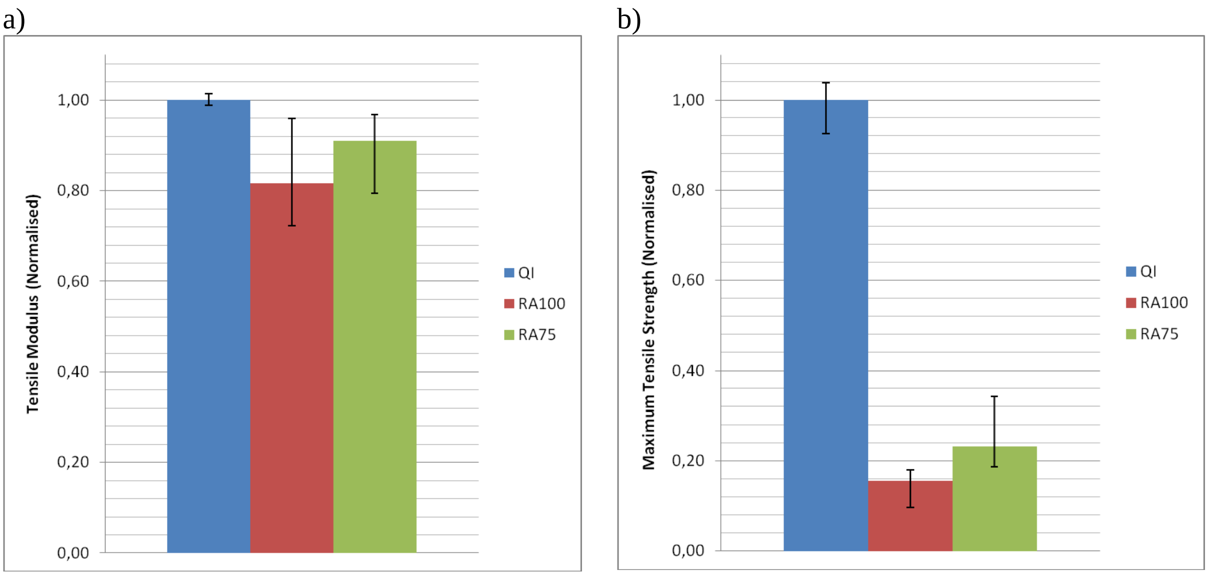

Figure 8. Tensile properties a) Modulus, b) Maximum tensile strength.

The tensile strength of the recycled panels showed significantly lower values than the continuous QI panel (Figure 8b). One explanation is that the presence of the reinforcement in the form of aggregates in the recycled panels leads to the presence of resin-rich regions and also stress concentrations around the edges of the aggregates which contribute to the reduction of the tensile strength [14, 16, 17]. Furthermore, the important lost in tensile strength can be attributed to the fact that the failure of the recycled panels is dominated by the matrix and that no fibre failure was observed in the fracture faces as noticed in section 3.1. Analogous results were observed on chopped tape-reinforce composites [14].

While both recycled panels showed tensile strength significantly lower than the continuous QI panel, the tensile strength of the recycled panel with a charge ratio of 75% were higher (+49%) than the recycled panel with a charge ratio of 100%. One possible reason for the higher strength of the RA75 specimens could be the reduction of resin-rich regions. However, researchers who worked on chopped taped or discontinuous staggered inclusions reinforced composites showed that the strength is well predicted by a toughness-based criterion [16, 22, 23]. A simplified expression of the strength, σ_c, given by the toughness-based criterion is given in (Eq. 1) where E_c is the composites modulus, G_c mode II interlaminar fracture toughness, H the composites thickness and h an initial damage size.

$$\sigma_c = \sqrt{\frac{2(H-h)E_c G_c}{Hh}} \tag{1}$$

Since E_c G_c and H are assumed to be of the same order of magnitude for both RA100 and RA75 panels, the higher strength of the RA75 specimens could be attributed to a smaller initial damage size h as well. The value h in the RA100 panel can be in a first approach estimated as 1 mm, that is the thickness of the initial scrap laminate. Since the thickness of the manufactured recycled panels, H, is of 3mm, the initial damage size, h, is of the order of one third of H and explains the low strength values. However, in the case of the RA75 panel, the stretching and squeeze flow induced by the macroscopic flow causes the aggregate to deform during the mould filling and therefore reduces its thickness [11, 17], that is, the order of h, which can explain the higher strength value recorded as compared to the RA100 panel. It can be expected that thicker panels will lead to higher strength values.

4. CONCLUSIONS

An alternative route to the traditional mechanical recycling technique of thermoplastic composites was investigated. Production scrap of a glass/PP laminate were cut into rectangular aggregates and processed in bulk form by compression moulding. The mechanical properties of the recycled aggregate-reinforced panels were characterised through tensile tests. It was found that the tensile modulus of the recycled composite demonstrated values close to the equivalent QI continuous fibre reinforced laminate of the same composition. However, a noteworthy decrease in the recycled composite strength was observed. This substantial decrease in strength was attributed to resin-rich regions, stress concentrations at the border of the aggregates and initial damage size in the tensile specimens. At this stage, the recycled composites show potential for stiffness driven applications and thick parts. Further investigation of process parameters aiming at reducing resin-rich regions and initial damage size should be considered in the view of increasing the strength of the recycled composite.

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