Tensile and impact properties of chopped carbon fibre reinforced thermoplastics with multiple recycle and regenerate

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ABSTRACT – REZUMAT

Tensile and impact properties of chopped carbon fibre reinforced thermoplastics with multiple recycle and regenerate

The increasing use of high-value carbon fibre in composites is linked with increasing waste generation. A simple and feasible chopped/hot-press method was proposed for multiple recycled carbon fibre-reinforced thermoplastic composites. The effect of regeneration times on the tension and impact properties of carbon fibre-reinforced polypropylene thermoplastic composites was investigated experimentally. The results showed that the r1-CFRTP specimen decreased by 69.34% in tensile strength and 48.66 % in tensile modulus compared with v-CFRTP. However, its tensile properties were improved with the increase of regeneration times (before 3 times). The impact strength of r2-CFRTP and r3-CFRTP is 12.65%, 20.85% higher than v-CFRTP, while r1-CFRTP and r4-CFRTP are 8.02% and 7.06% lower than v-CFRTP. When the third regeneration makes relatively excellent mechanical properties for recycled carbon fibre/PP composite, the chopped/hot-press method is a meaningful attempt at recycling and reusing the thermoplastic composites.

Keywords: *chopped carbon fibre, multiple recycle, thermoplastic polymer, tensile, impact*

Proprietăți de rezistență la tracțiune și la impact ale termoplasticelor armate cu fibră de carbon tăiată, cu reciclare și regenerare multiplă

Utilizarea tot mai frecventă a fibrei de carbon cu valoare adăugată în compozite este legată de creșterea generării de deșeuri. O metodă simplă și fezabilă de presare la cald/tăiere a fost propusă pentru mai multe compozite termoplastice armate cu fibră de carbon reciclată. Influența timpilor de regenerare asupra proprietăților de rezistență la tracțiune și la impact ale compozitelor termoplastice din polipropilenă armate cu fibre de carbon a fost investigată experimental. Rezultatele au arătat că pentru proba r1-CFRTP, rezistența la tracțiune scade cu 69,34% și modulul de tracțiune cu 48,66%, în comparație cu proba v-CFRTP. Cu toate acestea, odată cu creșterea timpilor de regenerare (până la 3 ori), proprietățile sale de rezistență la tracțiune au fost îmbunătățite. Rezistența la impact a probelor r2-CFRTP și r3-CFRTP este 12,65%, cu 20,85% mai mare decât cea a probei v-CFRTP, în timp ce cea a probelor r1-CFRTP și r4-CFRTP este cu 8,02% și 7,06% mai mică decât cea a probei v-CFRTP. În timp ce cea de-a treia regenerare face ca proprietățile mecanice să fie relativ excelente pentru compozitul din fibră de carbon reciclată/PP, metoda prin presare la cald/tăiere este o încercare semnificativă de reciclare și reutilizare a compozitelor termoplastice.

Cuvinte-cheie: *fibră de carbon tăiată, reciclare multiplă, polimer termoplastic, tracţiune, impact*

INTRODUCTION

Carbon fibre reinforced polymer composite (CFRP) is used in a wide range of applications in the automotive [1], construction industries [2], and other engineering applications because of its superior specific strength and stiffness and lower density [3–6]. CFRP can be divided by the component matrix polymers as either carbon fibre-reinforced thermosetting composites (CFRP) or thermoplastic composites (CFRTP). CFRTP has been growing at high speed, especially in the civil field, such as sports equipment, and new energy vehicle applications [7]. The main reasons are their better recyclability and ability to be processed more rapidly compared with thermosetting based [1, 4, 8]. Moreover, CFRTP can be applied in construction parts where toughness is more important than strength and stiffness [9]. CFRP recycling approach can be summarised into three broad methods, mechanical, thermal, and chemical, which use a corresponding source to separate the carbon fibres from the thermoset matrix respectively [10]. Those recovery processes are complex as well as cause carbon fibre damage. A thermoplastic plastic is a kind of linear polymer material, which can realize solid-liquid conversion above the viscous flow temperature and resolidify with the decrease in temperature. Therefore, thermoplastic composites can be recovered using remelting and remodelling. In other words, CFRTP is more recyclable compared with thermosetting-based composites. Since thermoplastic-based composites can be shaped and formed repeatedly by heating and pressing, they can be developed to deal with the recycling and environmental pressure caused by the waste of thermosetting-based composites.

There are continuous and discontinuous filling forms of carbon fibre in CFRTP, a drawback of continuous CFRTP is the limited formability. Discontinuous fibre shows superiority in complex shape design, which has been widely prepared using compression moulding, injection-moulded, and pultrusion processes, these processes may ensure the fabrication of discontinuous CFRTP with recycled carbon fibres. At present, increased work is conducted on this type of recycling for discontinuous fibre-reinforced thermoplastic composites because the recycled carbon fibres for low-end fields are step-by-step. Carbon fibre sheet moulding composite produced via chopped carbon fibre and thermoplastic plastic hot-press moulding approach presents a balanced solution with excellent mechanical performance and better formability [11]. Yi Wan [11, 12] comprehensively studied the preparation process and influence factors of semi-prepreg carbon fibre chopped tape reinforced thermoplastic composites, a wet type of paper-making method was used for better dispersion. The tape fibre lengths, thickness and moulding pressures have an impact on the mechanical properties. Liu [13] proposed a N-methyl-2-pyrrolidone solution impregnation method to prepare recycled carbon fibre-reinforced Polyetherimide chopped tapes. It is not necessary to dissolve all resin in the fabric thus has improved recovery efficiency.

It is an effective strategy to make full use of resources and save costs by multiple recycling of carbon fibre. Longana [14] adopted resin burning (pyrolysis) as a carbon fibre reclamation process, hot pressure, and vacuum bag moulding as remanufacturing processes and investigated the performance of short carbon fibre reinforced epoxy resin composite after recycling twice. The recovery of carbon fibre was highly oriented by wet suspension and pressure injection. The experimental results show that reduced adhesion between the fibre and matrix caused by residues builds up on the fibre surface during the pyrolysis process. Concerns have been raised regarding the degradation of recycled carbon fibre during the resin removal process. It is evident that the current thermoset material paradigm is not conducive to closed-loop composite recycling [15]. Tapper [15, 16] evaluated a closed-loop recycling methodology for discontinuous carbon fibre-reinforced thermoplastic matrix through a dissolution/precipitation reclamation technique and remanufacturing with the Hi-PerDiF method.

The purpose of this investigation is to determine if thermoplastic composite is multiple recyclables based on the principle of linear polymer melt remodelling. We explored the multiple recovery process of carbon fibre-reinforced polypropylene. The recycle chopped carbon fibre tape is prepared into prepreg by moulding, before that, chopped tapes were vibrated by a modified oscillator to achieve better dispersion and uniformity. The recycled chopped CFRTP can be used as other discontinuous thermoplastic composites. For instance, application in a cover plate of a vehicle trunk, electrical components, or sports helmet. These new processing steps were introduced to address both the performance and processing issues. The effect of recovery times on fibre morphology, distribution and mechanical properties of each stage was discussed.

EXPERIMENTAL

Materials

Polypropylene was supplied by Shanghai Petrochemical Co., Ltd., China, and its melt index is 26 g/10 min. Carbon fibre plain fabric was supplied by Henan Yongmei Carbon Fibre Co., Ltd., China.

Manufacturing of multiple recoveries CFRTP

1. Firstly, polypropylene pellets were prepared into thin films by hot pressing. Virgin CFRTP for the mechanical characterization and recycling were prepared by compression moulding. 180°C with a pre-pressing step of 20 min at 4 MPa and a pressing step of 6 hours at 2 MPa as the machine cooled naturally.
2. Virgin CFRTP were cut into a specified size (4×4 cm) before the recycling process for simulating the crushing process of waste. The chopped carbon fibre tapes were placed in a special mould to ensure uniformity with a vibrating machine. Prepreg of chopped carbon fibre tapes with 0.5 mm thickness was pressurized at 180°C, 50 s at 4 MPa. Then 4 pieces of prepreg were laminated and hot pressed to prepare the first recycled chopped CFRTP (r1-CFRTP).
3. The process of step 2 was repeated, r1-CFRTP was cut into the chopped carbon fibre tapes and manufactured to r2-CFRTP, four times recovery processes were conducted, v-CFRTP, r1-CFRTP, r2-CFRTP, r3-CFRTP, r4-CFRTP were papered. A schematic diagram of the chopped recycling technique is shown in figure 1.

The weighing method was used to calculate the carbon fibre volume fraction of v-CFRTP specimens, as shown in equation 1:

$$v_f = \frac{w_f / \rho_f}{L \times W \times H} \quad (1)$$

where v_f is the fibre volume fraction; w_f and ρ_f are the weight and density of carbon fibres; L, W and H are the length, width, and thickness of composite specimens. The combustion absorption method was used to calculate the carbon fibre volume fraction of regenerative according to the GB/T31292-2014 method.

Experiments

Tensile test

Tensile testing was carried out on grades using a universal mechanical testing instrument (WDW-20) in a standard laboratory atmosphere. The specimen size is 150 mm × 12.5 mm × 2 mm, and five specimens from each quality were tested, according to the

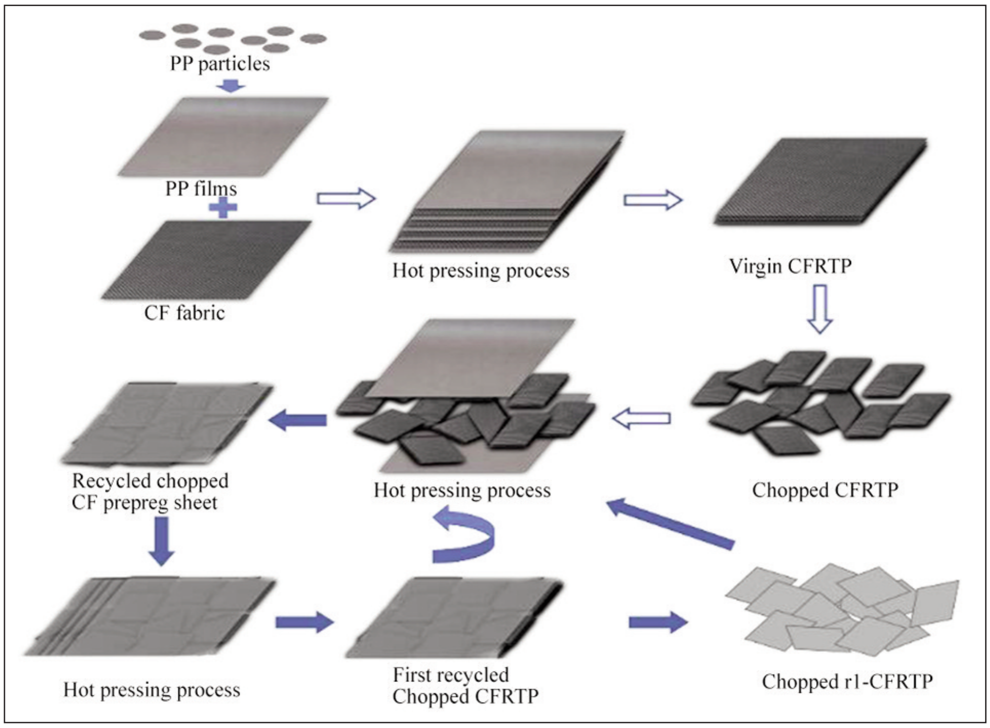

Fig. 1. A schematic diagram of multiple chopped melting recovery process

ASTMD3039 method, at a crosshead speed equal to 2 mm/min.

Notch impact test

The impact tests were carried out using a digital display simple cantilever impact tester (XJJ-50S). The absorbed energy during fracture in an impact test is used as a measure of impact strength. The dimensions of impact test specimens were about 50×10 mm (length and width). A notch was artificially prepared in the middle part of the specimens. Five specimens from each quality were tested.

The impact strengths of the specimen were calculated by the formula which references equation 2:

$$a = \frac{Ak}{W \cdot H} \times 10^3 \qquad (2)$$

where a is impact strength, which is available directly from the digital display. W, H, and A are the width of the specimen, the thickness of the specimen, and the absorbed energy of the specimen, respectively.

Morphology observation

The surface and edge of the specimen were performed to observe the microstructure of the specimen after testing using an electronic magnifying camera (Gaoping, GP-300C) aim to understand fibre arrangement and resin impregnation in the composite. The fracture surface of each specimen after the impact test was inspected and analysed using a scanning electron microscope (SEM, Hitachi, S-4800) after the impact test.

RESULTS AND DISCUSSION

Morphology analysis

Representative photographic images are shown in figure 2 for the surface and edge sides of all the tested specimens, figure 2, shows the vertical and horizontal interleaving structure shape of virgin carbon fibre plain fabric, from the edge side, the fibre waviness and layered structure are obvious due to the lamination process. r1-CFRTP still shows some characteristics of plain fabrics, that is, the arrangement of fibres is in some order although it has become chopped tapes. With the increase in recycling and regeneration times, the arrangement of fibres in the composite is becoming more and more disordered. Besides, it can be observed from the edge side image that the fusion situation of polypropylene resin in composite was more uniform for regenerated CFRTP.

Tensile performance

Representative stress-strain curves obtained by the tensile test of CFRTP for each stage are shown in figure 3. The specimens of virgin CFRTP showed linear-elastic tensile behaviour and brittle failure. The r-CFRTP responded linearly under tension initial stage, while they are not brittle like continuous carbon fibre reinforcement. A decrease in stiffness and failure properties can be observed for recycled specimens. The continuous unrecycled carbon fibre reinforcement has the best tensile performance. However, there is no necessarily positive proportional relationship between tensile properties and regeneration times, it is not that the more recycling times, the more tensile performance degradation.

Compared with unrecycled continuous carbon fibre, the tensile strength of the first recycled carbon fibre decreased by 69.34%, as shown in figure 4. It is obvious that continuous fibre becomes discontinuous ones, the tensile strength and modulus will be greatly reduced owing to the change in failure mechanism.

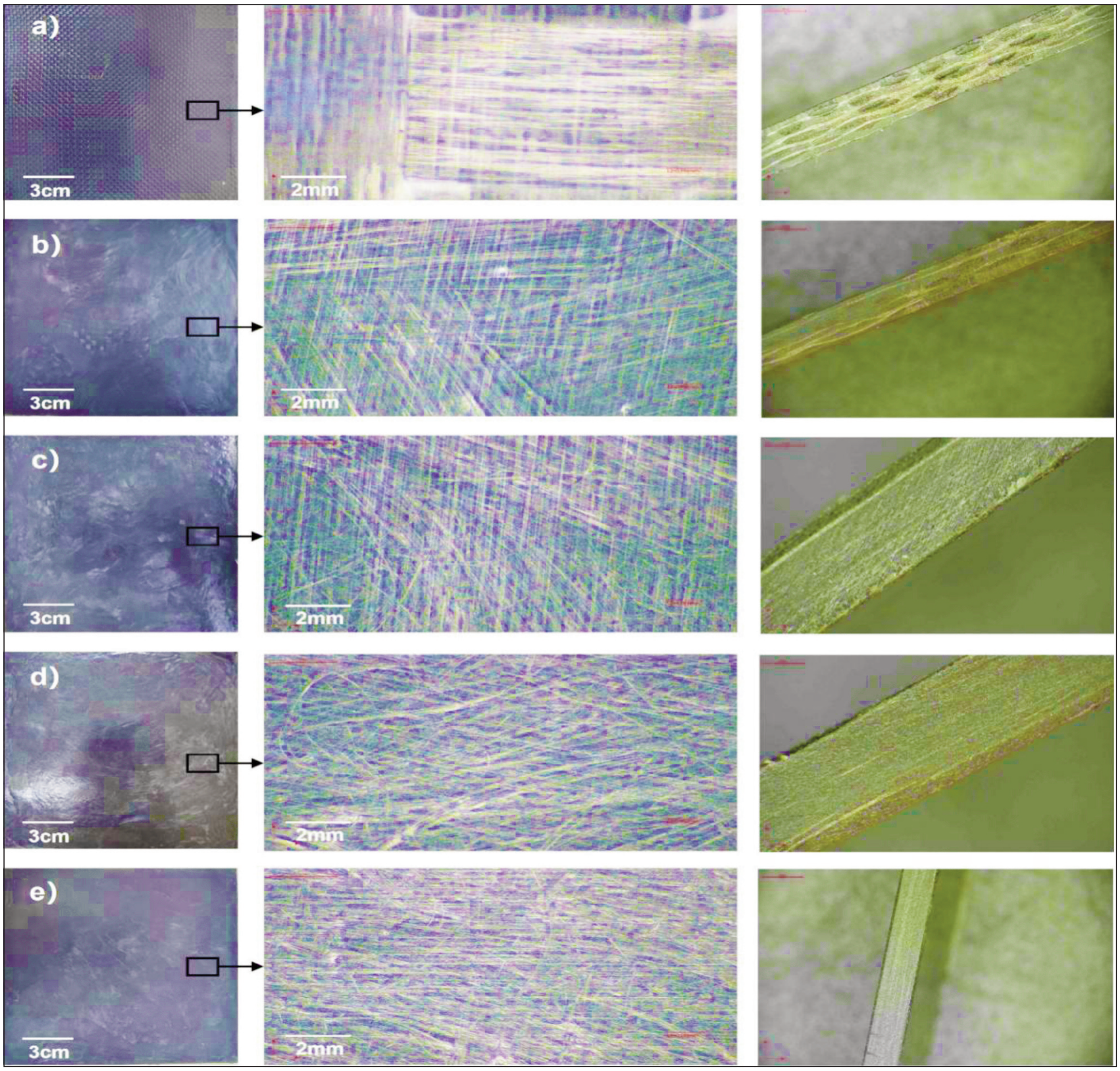

Fig. 2. Electronic photos of surface and edge for: *a* – CFRTP; *b* – r1-CFRTP; *c* – r2-CFRTP; *d* – r3-CFRTP; *e* – r4-CFRTP

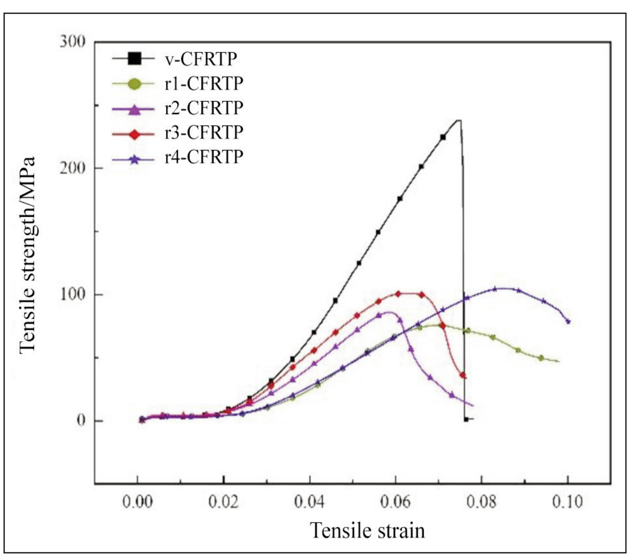

Fig. 3. Stress-strain curves for the tensile specimen

The tensile strength and modulus of second-generation recycled CFRTP are higher than that of r1-CFRTP. It may be the fibre waviness in-plane would decrease the strength performance. For sample r1-CFRTP, the fibre waviness of the fibric is still obvious, which can be observed in figure 2. Young's modulus and tensile strength are degraded seriously with increasing fibre waviness. The fibre waviness gradually decreased after the shearing and chopping process many times. The third-generation recycled CFRTP further improved. The different fibre length of the composite materials results in different mechanical properties, an equilibrium point was reached at the third recovery. The r3-CFRTP has the best tensile properties among recycled materials. Tensile failure was strain dominated, which suggests that the fibres failed by pull-out rather than at their cross-section (figure 5).

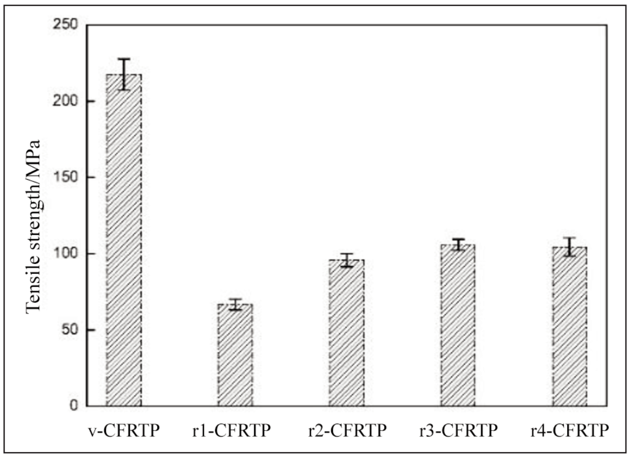

Fig. 4. Tensile strength of five specimens

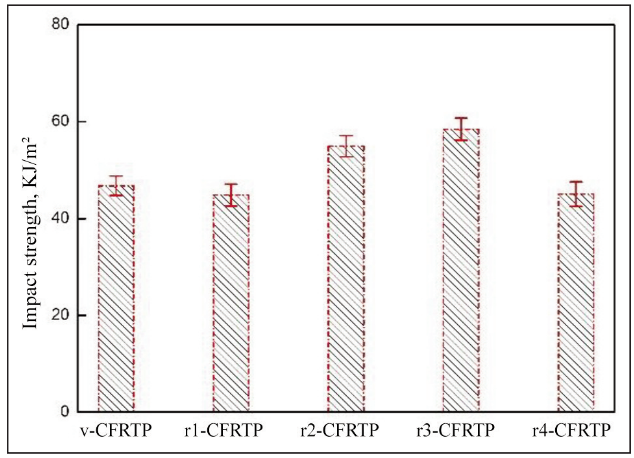

Fig. 6. Impact strength of five specimens

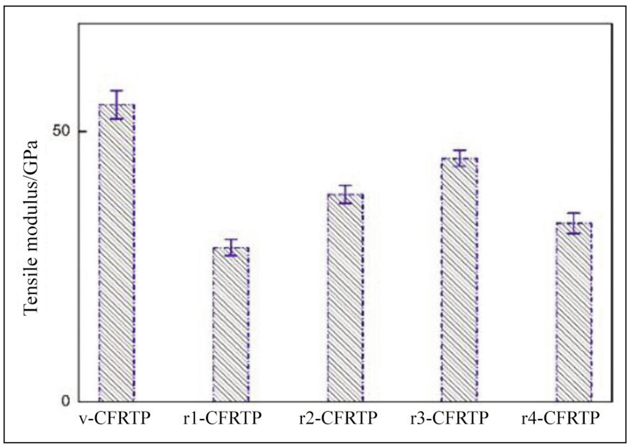

Fig. 5. Tensile modulus of five specimens

Impact performance

In general, impact phenomena are classified as low-velocity impact, high-velocity impact, ballistic impact, and hypervelocity damage impact [17]. In this investigation, the low-velocity impact carried on a cantilever beam tester was used to simulate the damage from dropped tools, bump, or fall during the services period. From figure 6, the impact strength of regenerated composites is even higher than that of virgin carbon fibre reinforced composite. The impact strength of r2-CFRTP and r3-CFRTP is 12.65%, 20.85% higher than v-CFRTP, while r1-CFRTP and r4-CFRTP are 8.02% and 7.06% lower than v-CFRTP. The results prove that the recovery and regeneration frequency is two or three will help to improve the impact properties of chopped tape CFRTP, which is of great significance for the utilization of waste carbon fibre-reinforced thermoplastic composites.

The duration of contact between the impactor and test sample is very high in the low-velocity impact process, and thus the sample absorbs energy by elastic deformation. In addition to the geometry of carbon fibre, other factors such as resin types and interfacial properties, also affect the impact properties [18]. The prime components of CFRTP are the carbon fibre, the matrix, and a fine interphase region, which determines the bonding strength between fibre and matrix. The properties of these individual components decide how the CFRTP elastic deformation and impact ruptures. Interphase plays a role to transfer stress between resin and reinforcement, whose properties are determined by the surface chemistry

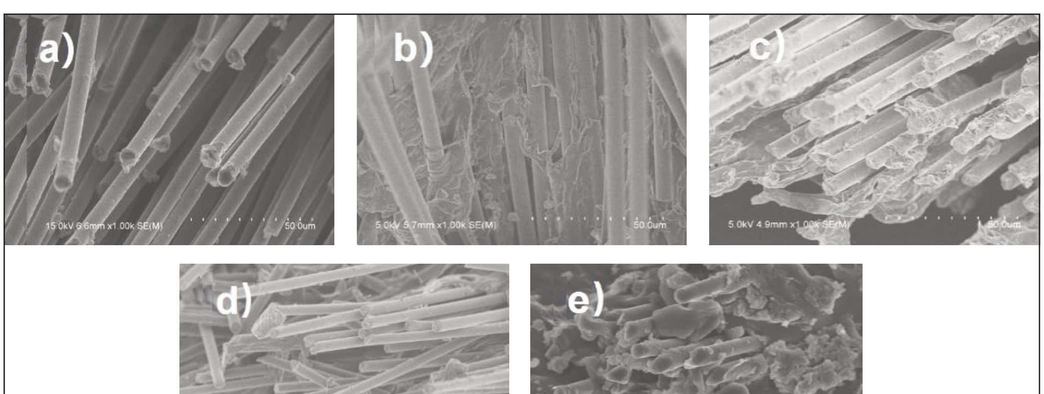

Fig. 7. Fracture surface of each specimen after impact: *a* – CFRTP; *b* – r1-CFRTP; *c* – r2-CFRTP; *d* – r3-CFRTP; *e* – r4-CFRTP

of fibres and resins and the composite material preparation process.

To further investigate the impact performance and fracture progress in the specimens, the x SEM picture of the impact section was observed. The obtained morphology results reveal that poor impregnation quality leads to a low impact strength for v-CFRTP. The changes in impregnation quality produce changes in interphase and interface strengths and stress transfer limitations. Besides, from figure 7, *d* and *e*, while at short carbon fibre length or poor impregnation quality, the failure is more likely to occur according to the interphase or interface debonding.

CONCLUSIONS

In this study, the effect of regeneration times on the tension and impact properties of carbon fibric reinforced polypropylene thermoplastic composites was investigated experimentally. From the tensile and notch impact test of the cantilever beam, and micrographic inspection, the following conclusions are reached:

- Multiple recycled chopped carbon fibre reinforced thermoplastics is a simple and feasible method for carbon and polymer resin circular and sustainable development.
- The r1-CFRTP specimen showed a total decrease of 69.34% in tensile strength and 48.66 % in tensile modulus compared with v-CFRTP. With the increase of regeneration times (before 3 times), its tensile properties were improved.
- The impact strength of r2-CFRTP and r3-CFRTP are 12.65%, 20.85% higher than v-CFRTP, while r1-CFRTP and r4-CFRTP are 8.02% and 7.06% lower than v-CFRTP. When the third regeneration makes relatively excellent mechanical properties for recycled carbon fibre/PP composite.

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