

Towards sustainable structural composites: A review on the recycling of continuous-fiber-reinforced thermoplastics

Alessandro Pegoretti

University of Trento, Department of Industrial Engineering, via Sommarive 9, 38123 Trento, Italy



ARTICLE INFO

Article history:

Received 5 December 2020

Received in revised form

29 January 2021

Accepted 4 March 2021

Keywords:

Thermoplastic composites

Continuous fibers

Recycling

Mechanical properties

ABSTRACT

In the last years, continuous-fiber-reinforced thermoplastic composites (CFRTCs) received an increasing interest for their potential advantages over composites with thermosetting resin mainly in terms of i) shorter manufacturing cycles ii) higher impact resistance and toughness and iii) recyclability at the end of first life. The main strategies for recycling CFRTCs are here reviewed on the basis of the information available in the scientific literature. The main recycling options currently available for CFRTCs can be broadly divided into mechanical, thermal and chemical methodologies. Moreover, the concept of all-polymer composites, i.e. composites in which both matrix and reinforcing components are made with thermoplastic polymers, is discussed in view of their relevance in recycling technology.

© 2021 Kingfa Scientific and Technological Co. Ltd. Publishing services by Elsevier B.V. on behalf of KeAi Communications Co. Ltd. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

1. Introduction

Polymer composites reinforced with continuous high-performance fibers have become an important class of structural engineering materials [1]. In fact, due to their high specific mechanical properties, they are extensively used in several applicative fields such as the automotive, aerospace, construction, and energy sectors [2]. The use of thermoplastics as matrix material in fiber reinforced composites has been growing steadily, especially for automotive and aerospace applications [3]. The main reasons behind the diffuse interest for CFRTCs are mainly based on their better recyclability and ability to be processed more rapidly than composites with thermosetting matrices [4,5]. Moreover, in comparison to thermosetting-based composites, CFRTCs also offer improved fracture toughness and impact strength [6] as well as the possibility to be easily joined using welding techniques [7]. Several thermoplastic polymers have been investigated as matrices for CFRTCs, including polyethylene (PE) [8–12], polypropylene (PP) [13–19], poly(ethylene terephthalate) (PET) [20], poly(butylene terephthalate) (PBT) [21,22], polyamide-6 (PA6) [23–29], polylactic acid (PLA) [30], polyurethane (PUR) [25], methacrylate (MA) [31], polyphenylene sulfide (PPS) [18,32,33], PA6 [34,35], PA66 [34], polyamide-11 (PA11) [15], polyetheretherketone (PEEK) [36–43], polyetherimide (PEI) [44–48], polyethersulfone (PES) [18,49] and

also liquid crystalline polymer (LCP) [50,51]. In CFRTCs, the above mentioned thermoplastic matrices are generally used to impregnate high-performances fibers such as carbon fiber (CF), glass fiber (GF), natural fiber (NF) and polymeric fibers in form of unidirectional tows or woven fabrics.

Incorporating fibers into high-viscous thermoplastic resins and achieving a good fiber wet-out are tasks much harder to be performed than in low-viscous thermosetting resins [1]. Nevertheless, several fiber incorporation techniques in thermoplastic resins have been developed, and many of them are now commercially used to produce thermoplastic preregs. The most common methods include i) hot-melt impregnation [52], ii) solution impregnation [53] iii) liquid impregnation [1], iv) film stacking [54], v) fiber mixing (i.e. comingled yarn) [55] and vi) dry powder coating [56]. Contrarily to thermosetting preregs, thermoplastic preregs can be stored for an unlimited period of time without any special storage facility and, whenever required, stacked and consolidated into laminates by the application of heat and pressure. Since thermoplastic matrix composites can be shaped and formed repeatedly by the application of heat and pressure, they can be processed using some metal-working as well as thermoplastic forming techniques, such as i) matched die forming [57], ii) hydroforming [58], and iii) thermoforming [59]. Moreover, tape placement is also becoming an important and widely used process for the manufacturing of CFRTCs [60]. Another emerging technique is liquid molding which has become an option with the availability of suitable reactive systems such as

E-mail address: alessandro.pegoretti@unitn.it.

Nomenclature			
η	viscosity	MA	methacrylate
ρ	density	NF	natural fibers
σ_b	stress at break	PA6	polyamide-6
ε_b	strain at break	PA66	polyamide-66
$\tan\delta$	loss tangent	PA11	polyamide-11
CF	carbon fiber	PE	polyethylene
CFRTC	continuous-fiber-reinforced thermoplastic composite	PEEK	polyetheretherketone
E	elastic modulus	PES	polyethersulfone
E'	tensile storage modulus	PET	poly(ethylene terephthalate)
EoL	end-of-life	PBT	poly(buthylene terephthalate)
FLD	fiber length distribution	PLA	polylactic acid
GF	glass fiber	PP	polypropylene
ILSS	interlaminar shear strength	PPS	polyphenylene sulfide
IS	impact strength	PVDF	poly(vinylidene fluoride)
LCP	liquid crystalline polymer	SFRTC	short fiber reinforced thermoplastic composites
M_w	weight average molecular weight	T_c	crystallization temperature
		T_g	glass transition temperature
		T_m	melting temperature
		X_c	crystallinity content

cyclic PBT [22], methacrylates (Elium) [31] and anionic polyamide-6 [61,62].

As illustrated in Fig. 1, during the preparatory operations and the manufacturing processes of CFRTCs there are several steps in which scraps can be generated. Therefore, with the increasing use of CFRTCs in several applicative sectors, the recovery and valorization of these scraps is becoming an environmental and economical challenge for the industries operating in this field. Moreover, the management of the CFRTCs components at the end of life first life also requires a special attention to the possibility of a full or at least partial recycling of the involved raw materials.

The currently available end-of-life (EoL) options for CFRTCs are schematically summarized in Fig. 2a. In landfilling, disposal of waste composites occurs in a sanitary landfill, constructed by lining an excavation with gravel, bituminous concrete and polyethylene sheet to create a barrier between the waste and surrounding environment [63]. From the environmental point of view this is certainly the worst option since no recovery is performed in terms of energy and raw materials. Moreover, landfilling introduces other negative burdens on the environment since i) landfill gas contains methane and carbon dioxide (both greenhouse gases), ii) contamination of soil and water can occur by toxic substances in the leachate, iii) land is diverted from other possible uses (such as agriculture) for future generations. A step toward a more

sustainable disposal of composites at the end of the first life is represented by incineration. In fact, through incineration part of the embodied energy can be recovered in the form of heat, with residues still going to landfill [64]. Options for a recovery of all or parts of the constituents of CFRTCs such as mechanical, thermal and chemical recycling processes, schematically represented in Fig. 2b, will be introduced and discussed in the following paragraphs.

With reference to the above scheme, the following main differences between thermoplastics vs thermosets based composites can be highlighted.

- Landfilling and incineration can be practiced for both thermoplastics and thermosets based composites.
- Mechanical recycling. Both thermoplastics and thermosets based composites can be shredded or crushed into particles or milled into fine powders. After this step, thermoplastics based composites can be reprocessed several times with the application of heat and pressure (see paragraph 2). On the contrary, thermosets based composites can only be used as fillers, reinforcement or raw materials for cement, concrete, among others [65].
- Thermal recycling can be practiced for both thermoplastics and thermosets based composites with no substantial differences.

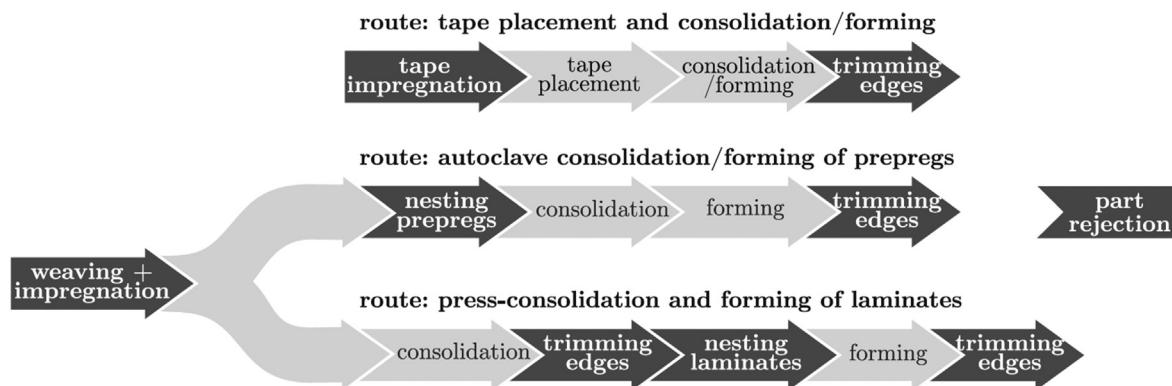


Fig. 1. Schematic overview of the main sources of scrap during the manufacturing of CFRTCs. The light-grey arrows represent the processing steps, whereas the dark-grey arrows highlight the operations where scrap is mainly generated. (reprinted from Vincent et al. [33] with permission of Elsevier).

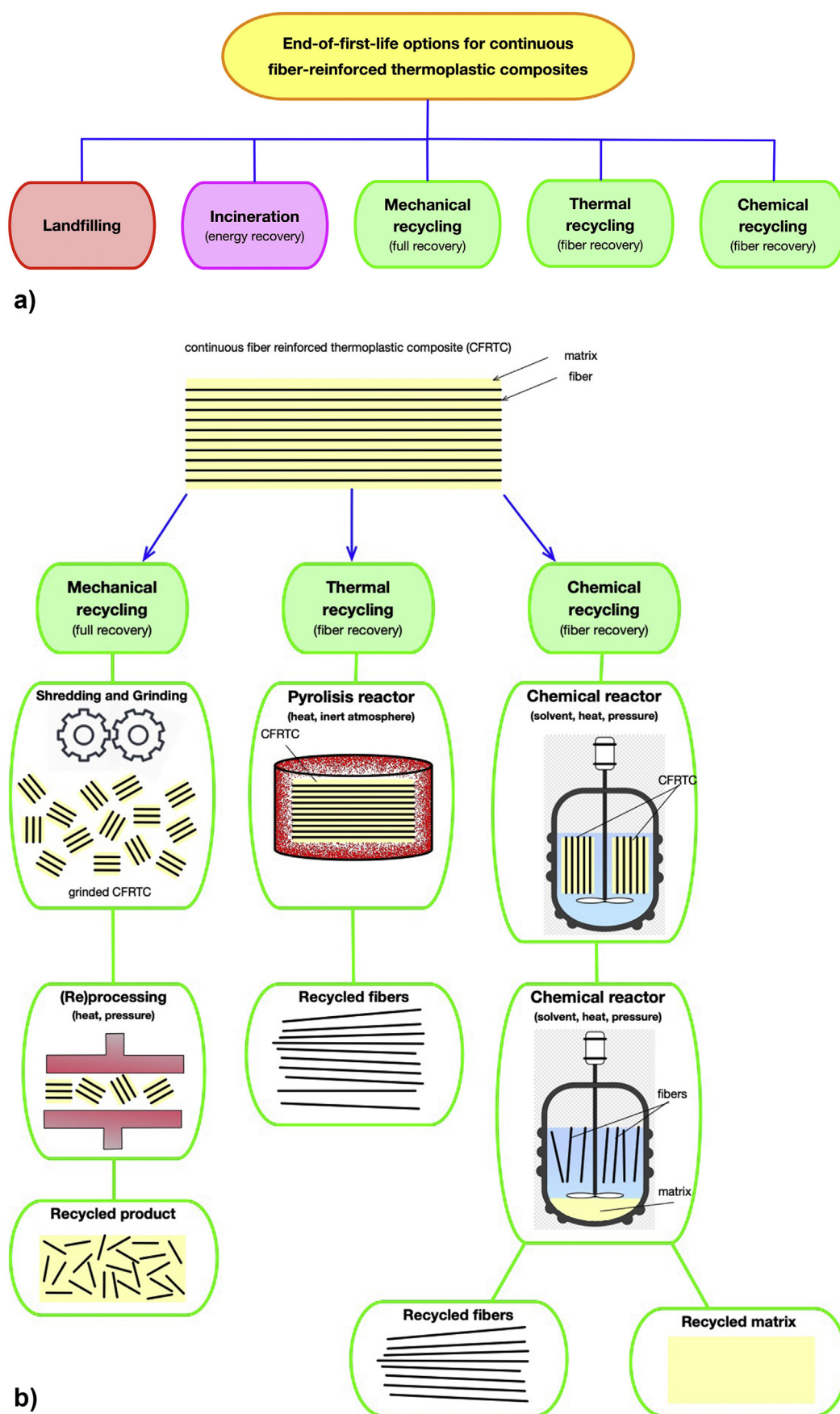


Fig. 2. a) End-of-life options for CFRTCs, b) main steps to move from the initial composite/scrap to the final recycled product for mechanical, thermal and chemical recycling.

- iv) Chemical recycling can also be practiced for both thermoplastics and thermosets based composites. Nevertheless, the chemical stability of thermosets is generally higher than those of thermoplastic polymers. Therefore, the recovery of fibers from thermosets based composites generally requires more drastic conditions. Moreover, it is important to mention that thermoplastic matrices can be dissolved and recovered from the solution, while thermosets matrices are generally degraded and cannot be therefore recovered.

2. Mechanical recycling

The first step in mechanical recycling of CFRTCs consist in reducing the size of the scrap off-cuts or of a component at its EoL by crushing or shredding followed by grinding [66]. The most adopted comminution techniques for composite parts make use of single [27] or multiple shaft shredding machines [18,33], of hammer mills or cutting mills [37]. The typical schematics of a multiple shaft shredder is reported in Fig. 3.

After shredding and grinding, recycled can be recovered and separated by sieving distinguishing between powders with a high resin content and fibers still embedded with resin. Due to the thermoplastic nature of the matrices used in CFRTCs, these fractions can be reprocessed by using conventional techniques such as compression molding (hot pressing) [18,21,26,33,36,67] extrusion-compression molding [21] or injection molding [15–17,21,22,26,27,31,37,68]. In some cases, due to their elevated fiber volume fraction, the viscosity of CFRTCs in the molten state is too high for injection molding. A dilution with neat resin is therefore required to lower the fiber content and the melt viscosity to the typical values of short fiber reinforced thermoplastic composites [15–17,22,27,32,37]. This often requires a melt compounding step in a twin-screw extruder or low-shear mixer [15] before the injection molding process. More recently also 3D-printing has been investigated as a possible re-processing route for CFRTCs [30].

Mechanical recycling unavoidably leads to a reduction of the length of the reinforcing fibers. Therefore, the recycled composite materials cannot longer be considered as CFRTCs but more properly as discontinuous fiber reinforced thermoplastic composites also called short fiber reinforced thermoplastic composites (SFRTCs). It is worthwhile to note that, for this class of composites, the fibers can efficiently increase the matrix modulus and strength only if a

good transfer of mechanical loads from the matrix to them is assured. All micromechanical models indicate that the maximum stress transferred to the fibers increases as the fiber length increases [69]. The maximum stress in the fiber can reach a limiting value corresponding to that of a unidirectional composite reinforced with the same volume fraction of continuous fibers and under the same applied external load. This limiting value can be reached provided that the fiber length reaches a minimum length called the “load transfer length” L_t [70]. Of course, the maximum stress in the fibers is also limited by their strength. Therefore, a critical fiber length, L_c , can be also defined as the minimum fiber length required to transfer to the fibers a maximum stress equal to its ultimate strength [70]. Therefore, fibers shorter than L_c are ineffective or even dangerous since they will be pulled out of the matrix under tensile load.

Of course all these consideration assume a perfect bonding at the fiber/matrix interface which is an unrealistic case [71]. A simply balance of the forces acting on a short fiber clearly indicate that the critical length depends on the fiber/matrix interfacial shear strength [69]. Therefore, in the mechanical recycling processes it is of crucial importance i) to preserve the fibers length at values higher than L_c [72] and ii) to assure that a suitable adhesion at the fiber/matrix interface is ultimately achieved [73,74]. It has been proven that a proper design of the screw profiles of extruders and of injection molding machines can contribute in reducing the intensity of the fiber breakage process [75].

Several investigations can be found in the scientific literature on the mechanical recycling (reprocessing) of CFRTCs. Some of the most relevant studies are listed in Table 1 where indications on the type of matrix materials (PP, PBT, PPS, MA, PA6, PA11, PLA, PEEK), the fiber type (CF, GF, NF, polymeric fibers), fiber content, the steps adopted for mechanical recycling along with the investigated materials properties are summarized.

In a study on PA6/GF CFRTCs, Moritzer and Heiderich [27] investigated the mechanical recycling of process-offcuts of continuous-fiber-reinforced composite sheets, consisting of a polyamide 6 matrix reinforced with a glass fiber fabric at 64 wt% content. After collection, the offcuts were shredded with a single-shaft shredder equipped with 18.5 kW drive power and a segment screen insert with holes having diameters of 8 mm and 10 mm. For the creation of pellets from the shredded material, a twin-screw extruder with a 25 mm screw and a ten barrel setup via

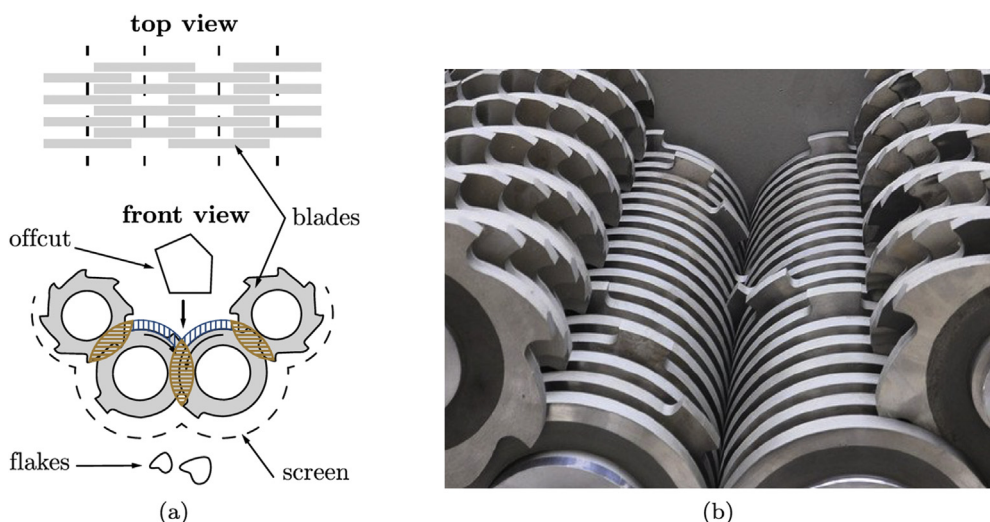


Fig. 3. Schematics of a multiple shaft shredder (four-shafts): a) top and front views; b) inside view of a four-shaft shredder (reprinted from Vincent et al. [33] with permission of Elsevier).

Table 1
Summary of the literature review on the mechanical recycling (re-processing) of CFRTCs.

Matrix	Fiber type/content	Steps for mechanical recycling	Measured quantities	Ref.
PP	NF/49 vol%	- Grinding + injection molding (up to 7 cycles)	E, σ_b , ε_b , η , T_c , X_c , FLD	[15]
PP	GF/60 wt%	- Shredding + twin-screw extrusion + injection molding	E, σ_b , ε_b , IS	[16]
PP	CF/31 vol%	- Shredding + twin-screw extrusion + injection molding	FLD, E, σ_b , IS, flammability,	[17]
PP	GF/not-reported	- Hot pressing	E, σ_b	[26]
PP	NF/50 wt%	- Grinding + injection molding (up to 3 cycles)	E, σ_b , IS	[68]
PBT	GF/35 wt%	- Grinding + reuse	E, σ_b , ε_b , IS, FLD, η	[21]
		- Grinding + injection molding	E, σ_b , ε_b , IS, FLD, M_w , X_c	
		- Grinding + extrusion-compression molding		
PBT	GF/58.7 wt%	- Grinding + compression molding		[22]
PBT	GF/58.7 wt%	- Grinding + injection molding	E, σ_b , ε_b , M_w , X_c , T_m	
PPS	CF/not-reported	- Shredding, low-shear mixing, compression molding	FLD, ρ , visual inspection	
PPS	CF/50 wt%	- Shredding + low shear mixing	E, σ_b	[32]
PPS	CF/not-reported	- Grinding + extrusion-compression molding	T_g , T_m , dynamic mechanical response	[67]
MA (Elium)	GF/22 wt%	- Thermoforming	E, σ_b	[31]
PA6	GF/67 wt%	- Grinding + injection molding		[27]
PA11	NF/50 vol%	- Shredding + twin-screw extrusion + injection molding	E, σ_b	
PLA	CF/not-reported	- Grinding + injection molding (up to 7 cycles)	E, σ_b , ε_b , η , T_c , X_c , FLD	
PEEK	CF/not-reported	- 3D-printing + re-manufacturing by 3D printing	E, σ_b , IS, M_w	[30]
PEEK	CF/not-reported	- Hammer milling + shredding + compression molding	E, σ_b , G' , $\tan\delta$, double lap shear strength	[36]
PEEK	CF/30,40,50 wt%	- Hammer milling + shredding + injection molding	E, σ_b , ε_b	[37]

the main feed was used. Moreover, the recycled PA6/GF composites were diluted with virgin PA6 added during the melt-compounding process through a side-feed along the barrel to reach fiber contents of 40, 30, 20 and 0 wt%. Finally, the obtained PA6/GF pellets were used to feed an injection molding machine with a screw diameter of 25 mm and a length-to-diameter ratio of 24 to produce samples for the mechanical tests. For comparison, also conventional virgin pellets of PA6 filled with 60 wt% of short glass fibers were used to produce samples by injection molding. The results in terms of tensile modulus and strength are summarized in Fig. 4. First of all, it is worthwhile to note that the mechanical properties of the pure recycled CFRTC material (i.e. without dilution with virgin PA6) are comparable with those of a conventional virgin PA6/GF short fiber reinforced composite with a similar fiber content. In fact, only a slight reduction of tensile modulus and strength can be noticed. Moreover, Fig. 4 clearly shows how the addition of recycled CFRTC to pure polyamide results in a remarkable increase in both the Young's modulus and the tensile strength.

Also reactive thermoplastic matrices have been investigated for the preparation of CFRTCs in light of their possible mechanical recyclability [22,31]. Steenkamer and Sullivan [22] explored the mechanical recycling of virgin composites prepared by liquid molding of a cyclic PBT resin and a knitted glass fabric with a fiber content of 58.7 wt%. In particular, the investigated CFRTC was recycled by a grinding/compounding/injection molding process. Prior to re-processing the ground cyclic composite by injection molding, it was dry blended with neat, virgin PBT to lower the reinforcement level to approximately 30 wt%. A variety of physical and mechanical tests were then conducted on the composite samples and compared with a baseline, commercially available short fiber reinforced PBT/glass composite. The values of modulus and strength obtained under various loading modes (tension, flexure, compression) are summarized and compared in Fig. 5. Interestingly enough, the mechanical properties of virgin and recycled systems are quite similar to those of the baseline material, with similar or only slightly lower modulus and strength values

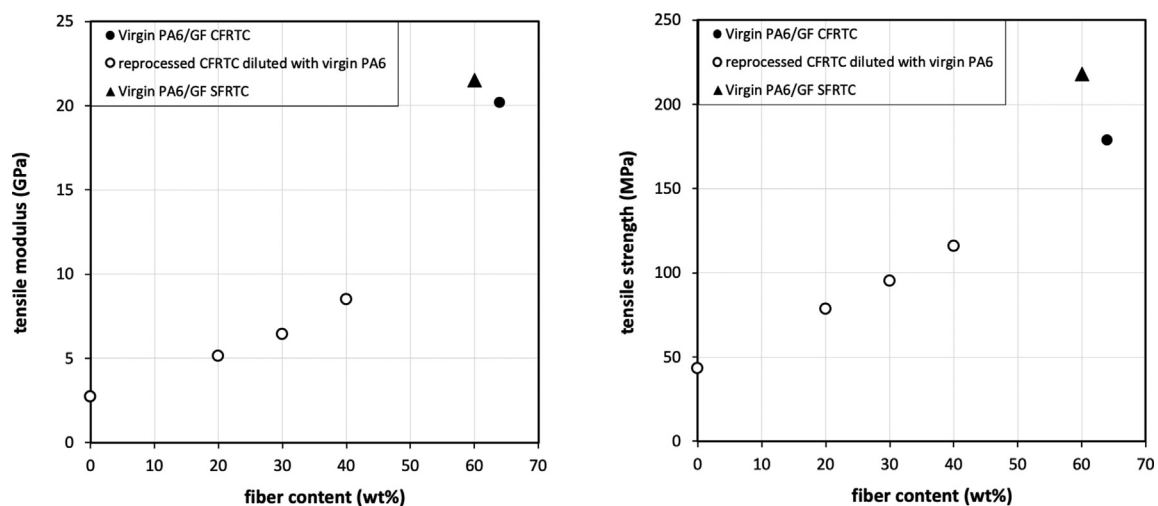


Fig. 4. Tensile modulus and strength of virgin PA6/GF CFRTC, reprocessed PA6/CFRTC diluted with virgin PA6, and short fiber reinforced PA/GF thermoplastic composites (data from Moritz et al. [27]).

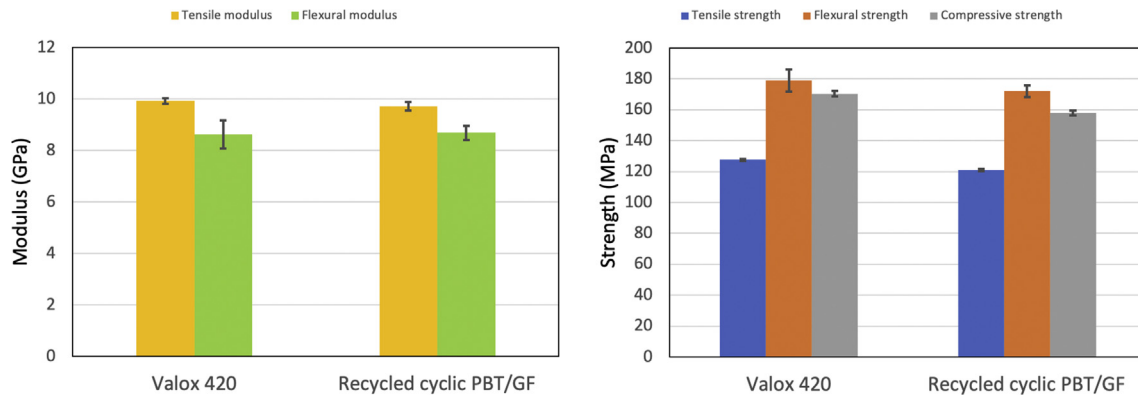


Fig. 5. Modulus and strength under various loading modes for virgin Valox 420 PBT/GF SFRTC and recycled cyclic PBT/GF CFRTC (data from Steenkamer and Sullivan [22]).

measured on recycled cyclic PBT/GF CFRTC in comparison to virgin Valox 420 PBT/GF SFRTC.

The authors also evaluated the stability of the investigated materials after soaking them in water at room temperature for various periods of time [22]. As reported in Fig. 6, the effects of the immersion in water were quite similar on both virgin Valox 420 PBT/GF SFRTC and recycled cyclic PBT/GF CFRTC. It is worthwhile to note that for both composites most of the degradation occurred within the first 3 days of exposure. Among the differences between the recycled and the baseline systems, it is worth mentioning that after 3 days of exposure the elongation at break of the Valox 420 dropped approximately by 10% while the elongation of the recycled cyclic dropped by almost 20%. This difference could be more likely attributed to a degradation of the fiber/matrix adhesion occurring in the recycled cyclic composite. The recyclability of PBT-based continuous thermoplastic composites reinforced with glass fibers was also investigated by Chu and Sullivan [21]. In particular, the authors concluded that after treatment with a suitable silane, processed regrind composites can be successfully recycled, with mechanical properties as good as those of commercial short fiber composites with the same composition.

A procedure for recycling PP-based sandwich panels was assessed by Corvaglia et al. [14]. In particular, they investigated sandwich panels with PP/GF laminates in the skins and PP foam as the core material. The panels were first ground by a mill and homogenized in a mixer or an extruder, then the pellets were re-ground and injection molded. A continuum increase of the tensile modulus was observed with the fiber content, while the tensile strength manifested a maximum for about 40 wt% of fibers. A drop of the Charpy impact strength was observed for the composites in comparison to the neat PP matrix [14].

Li and Englund [36] demonstrated that post-industrial trimmings and off-cuts of PEEK/CF composite can be successfully recycled into new composite products. In particular, after comminution via hammer mill and/or shredder, the recycled PEEK/CF materials were compression molded into flat panels whose mechanical properties resulted to be dependent on the size of the shredded fraction. The manufactured flat panels were also post thermoformed thus proving the possibility of a closed-loop recycling.

The concept of mechanical recycling and remanufacturing of CFRTC has been also recently investigated on 3D printed continuous carbon fiber reinforced PLA composites. Tian et al. [30] proved that PLA impregnated carbon fiber filament from 3D printed composite components can be recycled and reused as the raw material for further 3D printing process.

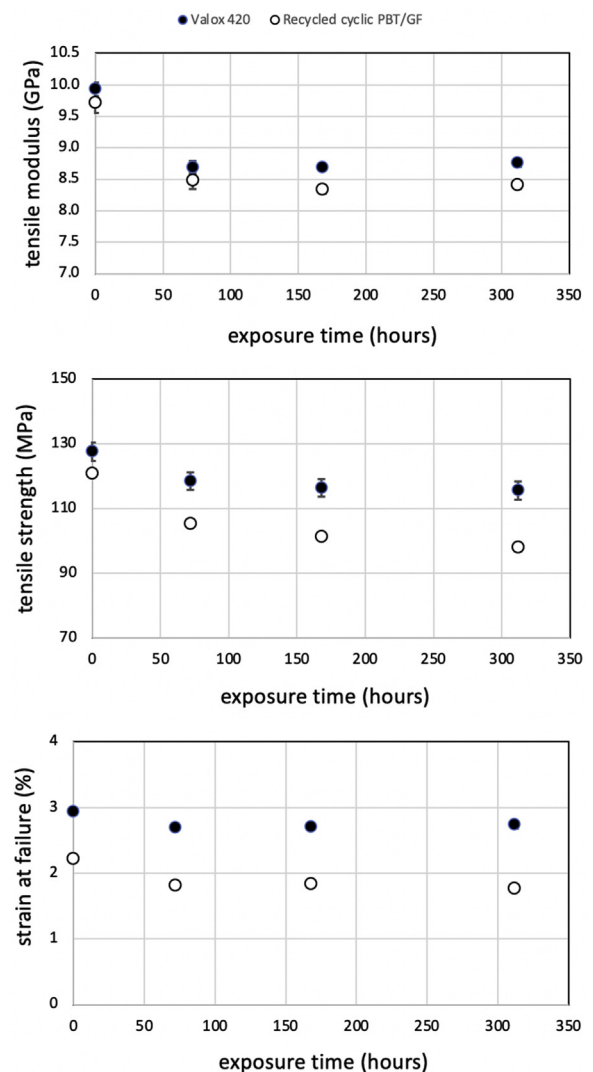


Fig. 6. Effect of exposure time in distilled water soak at room temperature on the modulus, strength and strain at failure of virgin Valox 420 PBT/GF SFRTC and recycled cyclic PBT/GF CFRTC (data from Steenkamer and Sullivan [22]).

An interesting concept that could open new possibilities in the recycling of CFRTCs is represented by the so called “all-polymer composites”. This class of composites includes i) single-polymer

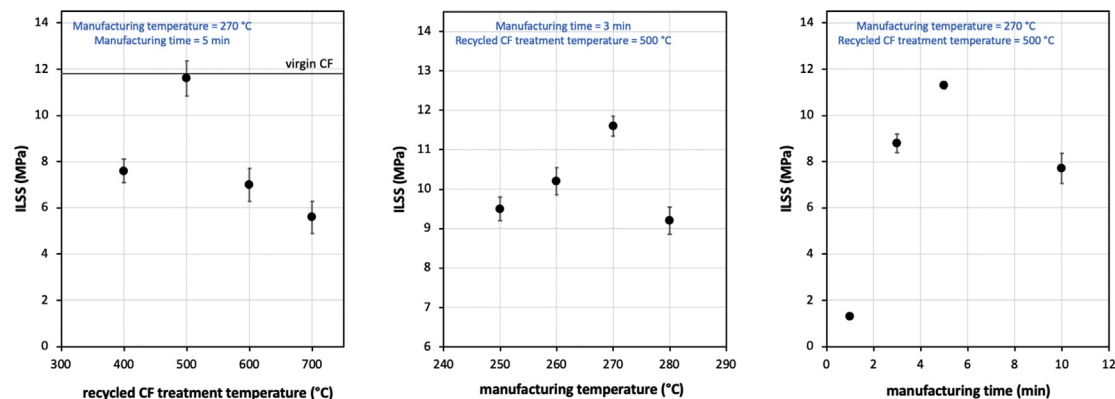


Fig. 7. Effect of the recycled carbon fiber treatment temperature during the pyrolysis process and of composite processing conditions (manufacturing temperature and manufacturing time) on the interlaminar shear strength values of the obtained recycled PET/CF composites (data from Baek et al. [29]).

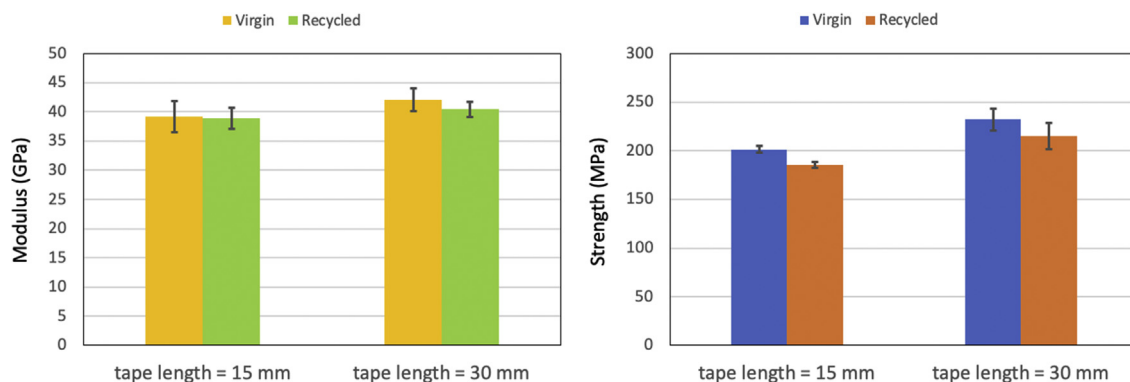


Fig. 8. Effect of chopped tape length on the tensile modulus and strength of virgin and recycled PEI/CF composites (data from Liu et al. [48]).

composites (SPC) in which the matrix and the reinforcement are of the same chemical composition, i.e. they are one-component systems, and ii) polymer-polymer composites (PPC) in which both matrix and reinforcing components are made with polymers, in some cases with the same chemical nature [76–85]. Several types of all-polymer composites have been investigated with polymers such as PE [8–10,79,86,87], PP [13,79,88–91], PET [87,92–94], PA6 [23,87,95], PLA [96], PVDF [97] and liquid crystalline polyesters [50,51]. At the end of the first life these composites can be remelted with a full recovery of the polymers they are made of [80,98,99]. Barany et al. [99] investigated the possibility of a re-processing of self-reinforced PP composites prepared by compression molding. In particular, the investigated composites consisted of sheets of a plain woven fabric of highly stretched split PP yarns as reinforcement and three kinds of PP as matrix material (β form of isotactic PP homopolymer, random PP copolymer, and a β form of the latter). Composite sheets with a thickness of 2.5 mm and a nominal reinforcement (i.e. α -PP fabric) content of 50 wt% were shredded, then extruded five times with a twin screw extruder and then granulated and injection molded after the first and fifth cycle in order to investigate the behaviour of the material during reprocessing [99]. On the basis of the results obtained from the thermo-mechanical characterization, the authors concluded that self-reinforced PP composites can well be reprocessed without any loss of properties of the matrix materials. The cyclic reprocessing (once and five times) did not result in considerable deterioration of the mechanical properties.

3. Thermal recycling

Thermal recycling is generally performed mainly to remove the polymer matrix of composites by thermal treatments with the aim to recover the fibers and reuse them again for the fabrication of composite materials. Methods for thermal recycling include pyrolysis, fluidized-bed pyrolysis and pyrolysis assisted with microwaves [66]. The above-mentioned techniques allow a recovery of the fibers and fillers while the matrix is generally volatilized into lower-weight molecules and gases such as carbon dioxide, hydrogen and methane for example, and an oil fraction, but also char on the fibers. Most processes operate at temperatures between 450 °C and 700 °C depending on the resin. The low temperature range is suitable for unsaturated polyester resin while high-performance thermoplastics, such as like PEEK for example, require higher temperatures [66]. The exposure to the elevated temperatures required for the pyrolysis processes can be detrimental for the mechanical performance of the recovered fibers [66,100].

Since mechanical recycling is not applicable to thermosetting composites, thermal and chemical recycling are often the only options for a partial material recovery from these composites. Therefore, most of the studies on the thermal recycling available in the scientific literature are mainly dealing with thermosetting composites [101–106] and only limited information are available on the thermal recycling of CFRTCs [29]. Baek et al. [29] used pyrolysis at 400 °C, 500 °C, 600 °C and 700 °C to obtain carbon fibers

from waste PA/CF composites. The recovered carbon fiber mats were used in combination with a PET matrix coming from recycled bottles and converted into film. To prepare PET/CF composites, the authors alternately stacked these PET films and carbon fiber mats followed by hot pressing at 270 °C for 5 min. The properties of the recycled PET/CF composites were compared with those of composites obtained with virgin CF. The interlaminar shear strength (ILSS) values of recycled PET/CF composites obtained under different processing conditions (manufacturing temperature and manufacturing time) by using recycled carbon fiber obtained with various treatment temperatures during the pyrolysis process are summarized in Fig. 7.

It is interesting to observe that for an optimized recycled CF treatment temperature of 500 °C the ILSS values approached those of composites obtained with virgin fibers (11.8 ± 0.8 MPa). Moreover, it can be noticed that the processing conditions can be optimized to maximize the ILSS values (in the specific case for a manufacturing temperature of 270 °C and a manufacturing time of 3 min) [29].

4. Chemical recycling

Chemical recycling of both thermosetting and thermoplastic polymeric composites consists in the removal of the matrix by solvolysis or dissolution in a proper solvent. In general, solvolysis implies the use of reactive solvents to break the covalent bonds of a thermosetting matrix, while dissolution refers to the phenomenon of dissolving thermoplastic polymer chains into a solvent, which is a purely physical process. The advantages of chemical recycling over thermal recycling, is that lower temperatures are generally required to degrade the polymeric matrices [66] with lower damages on the recovered fibers. In fact, dissolution and solvolysis can be operated by using a wide spectrum of solvents in a range of temperatures and pressure, with various catalysts. Moreover, contrarily to thermal recycling, chemical recycling by matrix dissolution allows a recovery of both the polymer matrix and full-length fibers [31].

Several investigation have been conducted on the dissolution or solvolysis of thermoplastic polymers of relevance for CFRTs such as PE [107], PA66 [108], PET [109], PBT [110] and PEEK [111]. Dissolution and solvolysis processes can be conducted i) at low temperature or ii) under supercritical conditions [112]. Among the investigated chemical solvents it is worthwhile to mention water (i.e. hydrolysis) [108,113,114], methanol (i.e. methanolysis) [110] and glycols (i.e. glycolysis [109,115], alcohols (i.e. alcolysis) [116], chloroform [31], an organic solvent (such as N-methyl-2-pyrrolidone) [48] or strong acids such as nitric acid [117,118].

A limited number of studies are available in the scientific literature on the chemical recycling of CFRTs [24,25,31,48]. For example, Liu et al. [48] investigated on the chemical recycling of CFRTs based on polyetherimide reinforced with carbon fibers. In particular, discarded PEI/CF composites were chopped in tapes (at two different length of 15 and 30 mm) and put in an organic solvent (N-methyl-2-pyrrolidone) for 2 h in order to dissolve the polymer matrix. The obtained chopped tapes were randomly placed in a container and shaken for 10 min in a shaker machine to obtain a uniform distribution. The obtained resin solution was filtered to separate any insoluble ingredients, some virgin resins was added and then poured into the container with uniformly distributed chopped tapes. After complete impregnation of the chopped fabric tapes, the solvent was evaporated in an oven. Finally, the material was thermally treated in a hot-press at 0.12 MPa and 290 °C for 10 min to consolidate the composites. The effect of chopped tape length on the tensile modulus and strength of virgin and recycled PEI/CF composites is summarized in Fig. 8. It is worthwhile to note

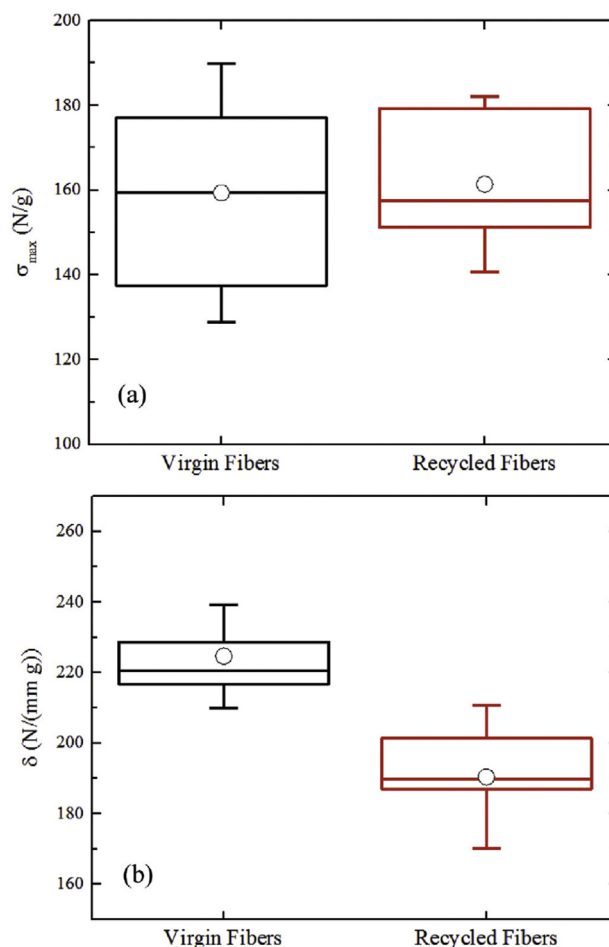


Fig. 9. Mass normalized tensile properties of glass fiber rovings recovered from dissolution of the thermoplastic part compared to those of virgin fibers. (a) Mass-normalized force at break (b) Mass-normalized slope of the load displacement curve, δ . The center points represent the mean, the center line the median, the box is the inter-quartile range, and the whiskers are 1.5 times the standard deviation. (reprinted from Cousins et al. [31] with permission of Elsevier).

that recycled composites present mechanical properties only slightly lower than virgin ones.

In the attempt to prove the possibility to recover the constituents of glass fiber thermoplastic composites from wind turbine blades, Cousins et al. [31] adopted a dissolution process on spar cap components made of MA(Elium)/GF by repeated soaking in fresh chloroform up to 72 h. After dissolution, the glass fibers were recovered and the polymer precipitated from the chloroform into methanol and dried. Tensile properties of the fibers recycled from the dissolution experiment were measured on rovings and compared with those of virgin fibers. The obtained results are reported in Fig. 9. It is interesting to observe that the mass-normalized strength of the fibers is preserved after the matrix dissolution process and that the mass-normalized stiffness shows only a limited (12%) reduction [31].

5. Conclusions and future trends

According to the analysis of the existing literature, it clearly emerges how recycling methods for CFRTC has been quite intensively investigated in recent years with the hope to effectively improve the circularity in materials usage. Most of the currently available CFRTC waste comes directly from production. As a matter

of fact, both mechanical and chemical recycling processes have been proven to allow a full recovery of the constituents (matrix and fibers) both from internal scraps and from products at the end of their first life.

Mechanical recycling appears to be the most suitable option for the recycling of CFRTC because it can be implemented with relatively simple techniques and it does not require the use of hazardous chemicals or high-temperature treatments. The main current limitation of mechanical recycling methods is the difficulty to preserve the fiber integrity and therefore to maintain adequate mechanical properties on the recycled materials.

Thermal recycling methods, such as pyrolysis, fluidized-bed pyrolysis and pyrolysis assisted with microwaves can also represent a recycling option for CFRTC. The main limitation in this case is that only the fibers are recovered while the thermoplastic matrix is completely degraded and removed during the process.

Chemical recycling methods, represented by dissolution or solvolysis of the polymer matrix, have also been quite actively investigated as recycling options for CFRTCs. In the case of matrix dissolution, both fibers and matrix can be fully recovered. The main limitations of chemical recycling is that they require a careful management of the solvents involved in the processes.

Future developments in the recycling opportunities for both CFRTCs can be forecasted in the following directions:

- i) an optimization of the parameters of mechanical recycling for a maximization of the fiber length after re-processing. This can be achieved though low-shear mixing conditions possible reached by an optimization of the profiles of the screws used in extrusion/injection molding processes;
- ii) a wider use of all-polymer composites, i.e. composites in which both matrix and reinforcing components are made with thermoplastic polymers, in some cases the polymers have the same chemical nature. In this case, mechanical recycling allows a full recovery of the constituents in form of polymer or polymer blends to be reused with processing technologies typical of thermoplastic materials;
- iii) an improvement of the chemical methods with the use of less harmful solvents such as water or alcohols in supercritical conditions to reach a matrix removal with a lower environmental impact.

Conflict of interest

None declared.

Acknowledgement

The author devotes this paper to the 85th anniversary of Prof. Stoyko Fakirov with the best wishes for a good health and further contributions to the polymer science.

References

- [1] P. Mallick, *Fiber-Reinforced Composites - Materials, Manufacturing, and Design*, third ed., Marcel Dekker, Inc, New York, USA, 2008.
- [2] E.J. Barbero, *Introduction to Composite Materials Design - third ed.*, CRC Press 2018.
- [3] M. Biron, *Thermoplastics and Thermoplastic Composites*, Elsevier, Amsterdam, The Netherlands, 2007.
- [4] S.-S. Yao, F.-L. Jin, K.Y. Rhee, D. Hui, S.-J. Park, Recent advances in carbon-fiber-reinforced thermoplastic composites: a review, *Compos. B Eng.* 142 (2018) 241–250.
- [5] R. Stewart, Thermoplastic composites — recyclable and fast to process, *Reinforc. Plast.* 55 (3) (2011) 22–28.
- [6] G. Jogur, A. Khan, A. Das, P. Mahajan, R. Alagirusamy, Impact properties of thermoplastic composites, *Textil. Prog.* 50 (3) (2018) 109–183.
- [7] T.J. Ahmed, D. Stavrov, H.E.N. Bersee, A. Beukers, Induction welding of thermoplastic composites—an overview, *Compos. Appl. Sci. Manuf.* 37 (10) (2006) 1638–1651.
- [8] A. Dorigato, G. Fredi, L. Fambri, J.-M. Lopez-Cuesta, A. Pegoretti, Polyethylene-based single polymer laminates: synergistic effects of nanosilica and metal hydroxides, *J. Reinforc. Plast. Compos.* 38 (2) (2019) 62–73.
- [9] M. Kazanci, D. Cohn, G. Marom, C. Migliaresi, A. Pegoretti, Fatigue characterization of polyethylene fiber reinforced polyolefin biomedical composites, *Composites Part A - Applied Science and Manufacturing* 33 (4) (2002) 453–458.
- [10] A. Pegoretti, M. Ashkar, C. Migliaresi, G. Marom, Relaxation processes in polyethylene fibre-reinforced polyethylene composites, *Compos. Sci. Technol.* 60 (8) (2000) 1181–1189.
- [11] H. Diaz-Alvarez, J.R. Picucci, M.H. McKenna, R.G. Lampo, Structural response of a recycled thermoplastic lumber bridge under civilian and military loads, *J. Thermoplast. Compos. Mater.* 28 (4) (2013) 461–478.
- [12] S.M. Khan, N. Gull, M.A. Munawar, A. Islam, S. Zia, M. Shafiq, A. Sabir, S.M. Awais, M.A. Butt, M.T.Z. Butt, T. Jamil, 2D carbon fiber reinforced high density polyethylene multi-layered laminated composite panels: structural, mechanical, thermal, and morphological profile, *J. Mater. Sci. Technol.* 32 (10) (2016) 1077–1082.
- [13] N. Cabrera, B. Alcock, J. Loos, T. Peijs, Proceedings of the institution of mechanical engineers, Part L, *J. Mater.: Design Appl.* 218 (2) (2004) 145–155.
- [14] P. Corvaglia, A. Passaro, O. Manni, L. Barone, A. Maffezzoli, Recycling of PP-based sandwich panels with continuous fiber composite skins, *J. Thermoplast. Compos. Mater.* 19 (6) (2006) 731–745.
- [15] C. Gourier, A. Bourmaud, A. Le Duigou, C. Baley, Influence of PA11 and PP thermoplastic polymers on recycling stability of unidirectional flax fibre reinforced biocomposites, *Polym. Degrad. Stabil.* 136 (2017) 1–9.
- [16] M.E. Otheguy, A.G. Gibson, E. Findon, R.M. Cripps, A.O. Mendoza, M.T.A. Castro, Recycling of end-of-life thermoplastic composite boats, *Plastics, Rubber Compos.* 38 (9–10) (2013) 406–411.
- [17] M. Tomioka, T. Ishikawa, K. Okuyama, T. Tanaka, Recycling of carbon-fiber-reinforced polypropylene prepreg waste based on pelletization process, *J. Compos. Mater.* 51 (27) (2017) 3847–3858.
- [18] G. Vincent, *Recycling of Thermoplastic Composite Laminates*, University of Twente, Enschede, the Netherlands, 2019.
- [19] M. Hou, K. Friedrich, Stamp forming of continuous carbon fibre/polypropylene composites, *Compos. Manufact.* 2 (1) (1991) 3–9.
- [20] J. Andrzejewski, M. Szostak, T. Bak, M. Trzeciak, The influence of processing conditions on the mechanical properties and structure of poly(ethylene terephthalate) self-reinforced composites, *J. Thermoplast. Compos. Mater.* 29 (9) (2016) 1194–1209.
- [21] J. Chu, J.L. Sullivan, Recyclability of a glass fiber poly(Butylene terephthalate) composite, *Polym. Compos.* 17 (3) (1996) 523–531.
- [22] D.A. Steenkamer, J.L. Sullivan, On the recyclability of a cyclic thermoplastic composite material, *Composites Part A* 29B (1998) 745–752.
- [23] Y. Gong, A. Liu, G. Yang, Polyamide single polymer composites prepared via in situ anionic polymerization of ϵ -caprolactam, *Compos. Appl. Sci. Manuf.* 41 (8) (2010) 1006–1011.
- [24] C. Chaabani, E. Weiss-Hortala, Y. Soudais, Impact of solvolysis process on both depolymerization kinetics of nylon 6 and recycling carbon fibers from waste composite, *Waste Biomass Valorization* 8 (8) (2017) 2853–2865.
- [25] F. Knappich, M. Klotz, M. Schlummer, J. Wolling, A. Maurer, Recycling process for carbon fiber reinforced plastics with polyamide 6, polyurethane and epoxy matrix by gentle solvent treatment, *Waste Manag.* 85 (2019) 73–81.
- [26] K. Kemmochi, H. Tkayanagi, C. Nagasawa, J. Takahashi, R. Hayashi, Possibility of closed loop material recycling for fiber reinforced thermoplastic composites, *Adv. Perform. Mater.* 2 (1995) 385–394.
- [27] E. Moritzer, G. Heiderich, *Mechanical Recycling of Continuous Fiber-Reinforced Thermoplastic Sheets*, 2016.
- [28] T. Ageyeva, I. Sibikin, J.G. Kovacs, A review of thermoplastic resin transfer molding: process modeling and simulation, *Polymers (Basel)* 11 (10) (2019).
- [29] Y.-M. Baek, P.-S. Shin, J.-H. Kim, H.-S. Park, D.-J. Kwon, K.L. DeVries, J.-M. Park, Investigation of interfacial and mechanical properties of various thermally-recycled carbon fibers/recycled PET composites, *Fibers Polym.* 19 (8) (2018) 1767–1775.
- [30] X. Tian, T. Liu, Q. Wang, A. Dilmurat, D. Li, G. Ziegmann, Recycling and remanufacturing of 3D printed continuous carbon fiber reinforced PLA composites, *J. Clean. Prod.* 142 (2017) 1609–1618.
- [31] D.S. Cousins, Y. Suzuki, R.E. Murray, J.R. Samaniuk, A.P. Stebner, Recycling glass fiber thermoplastic composites from wind turbine blades, *J. Clean. Prod.* 209 (2019) 1252–1263.
- [32] T.A. de Bruijn, G. Vincent, F.W.J. van Hattum, Recycling C/PPS Laminates into Long Fibre Thermoplastoc Composites by Low Shear Mixing, 21st International Conference on Composite Materials, Xi'an, China, 2017.
- [33] G.A. Vincent, T.A. de Bruijn, S. Wijskamp, M.I. Abdul Rasheed, M. van Drongelen, R. Akkerman, Shredding and sieving thermoplastic composite scrap: method development and analyses of the fibre length distributions, *Compos. B Eng.* 176 (2019).
- [34] E. Botelho, Mechanical behavior of carbon fiber reinforced polyamide composites, *Compos. Sci. Technol.* 63 (13) (2003) 1843–1855.
- [35] H. Li, Y. Wang, C. Zhang, B. Zhang, Effects of thermal histories on interfacial properties of carbon fiber/polyamide 6 composites: thickness, modulus, adhesion and shear strength, *Compos. Appl. Sci. Manuf.* 85 (2016) 31–39.

- [36] H. Li, K. Englund, Recycling of carbon fiber-reinforced thermoplastic composite wastes from the aerospace industry, *J. Compos. Mater.* 51 (9) (2016) 1265–1273.
- [37] G. Schinner, J. Brnadt, H. Richter, Recycling carbon-fiber-reinforced thermoplastoc composites, *J. Thermoplast. Compos. Mater.* 9 (1996) 239–245.
- [38] M. Roux, N. Eguémann, C. Dransfeld, F. Thiébaud, D. Perreux, Thermoplastic carbon fibre-reinforced polymer recycling with electrodynamic fragmentation: from cradle to cradle, *J. Thermoplast. Compos. Mater.* 30 (3) (2017) 381–403.
- [39] S. Ramakrishna, W.K. Tan, S.H. Teoh, M.O. Lai, Recycling of carbon fiber/peek composites, *Key Eng. Mater.* 137 (1997) 1–8.
- [40] G. Zhang, R.A. Latour Jr., J.M. Kennedy, H. Del Schutte Jr., R.J. Friedman, Long-term compressive property durability of carbon fibre-reinforced polyetheretherketone composite in physiological saline, *Biomaterials* 17 (1996) 781–789.
- [41] H. Luo, G. Xiong, K. Ren, S.R. Raman, Z. Liu, Q. Li, C. Ma, D. Li, Y. Wan, Air DBD plasma treatment on three-dimensional braided carbon fiber-reinforced PEEK composites for enhancement of in vitro bioactivity, *Surf. Coating. Technol.* 242 (2014) 1–7.
- [42] A. Almajid, K. Friedrich, J. Floeck, T. Burkhart, Surface damage characteristics and specific wear rates of a new continuous carbon fiber (CF)/polyetheretherketone (PEEK) composite under sliding and rolling contact conditions, *Appl. Compos. Mater.* 18 (3) (2010) 211–230.
- [43] K. Fujihara, Z.-M. Huang, S. Ramakrishna, H. Hamada, Influence of processing conditions on bending property of continuous carbon fiber reinforced PEEK composites, *Compos. Sci. Technol.* 64 (16) (2004) 2525–2534.
- [44] M. Lu, L. Ye, Y.-W. Mai, Thermal de-consolidation of thermoplastic matrix composites—II. “Migration” of voids and “re-consolidation”, *Compos. Sci. Technol.* 64 (2) (2004) 191–202.
- [45] M. Sharma, I. Mohan Rao, J. Bijwe, Influence of fiber orientation on abrasive wear of unidirectionally reinforced carbon fiber–polyetherimide composites, *Tribol. Int.* 43 (5–6) (2010) 959–964.
- [46] M. Sharma, I.M. Rao, J. Bijwe, Influence of orientation of long fibers in carbon fiber–polyetherimide composites on mechanical and tribological properties, *Wear* 267 (5–8) (2009) 839–845.
- [47] G. Xian, Z. Zhang, Sliding wear of polyetherimide matrix composites, *Wear* 258 (5–6) (2005) 776–782.
- [48] B. Liu, P. Zhu, A. Xu, L. Bao, Investigation of the recycling of continuous fiber-reinforced thermoplastics, *J. Thermoplast. Compos. Mater.* 32 (3) (2019) 342–356.
- [49] G.M. Wu, J.M. Schultz, Processing and properties of solution impregnated carbon fiber reinforced polyethersulfone composites, *Polym. Compos.* 21(2) pp. 223–230.
- [50] A. Pegoretti, A. Zanolli, C. Migliaresi, Preparation and tensile mechanical properties of unidirectional liquid crystalline single-polymer composites, *Compos. Sci. Technol.* 66 (13) (2006) 1970–1979.
- [51] A. Pegoretti, A. Zanolli, C. Migliaresi, Flexural and interlaminar mechanical properties of unidirectional liquid crystalline single-polymer composites, *Compos. Sci. Technol.* 66 (13) (2006) 1953–1962.
- [52] B. Hayes, J. Seferis, The effect of fabric tension and the number of impregnation rollers on woven fabric prepreg quality and cured laminates, *Composites Part A* 28A (1997q) 791–799.
- [53] K.E. HGoodman, Thermoplastic Prepreg Manufacture by Solution Impregnation, Virginia Polytechnic Institute and State University, Blacksburg, Virginia, 1990.
- [54] W.J.B. Grouve, R. Akkerman, Consolidation process model for film stacking glass/PPS laminates, *Plastics, Rubber Compos.* 39 (3–5) (2013) 208–215.
- [55] N. Bernet, V. Michaud, P.-E. Bourban, J.-A.E. Manson, Commingled yarn composites for rapid processing of complex shapes, *Compos. Appl. Sci. Manuf.* 32 (2001) 1613–1626.
- [56] S.R. Iyer, L. Drzal, Manufacture of powder-impregnated thermoplastic composites, *J. Thermoplast. Compos. Mater.* (1990) 325–355.
- [57] J. Krebs, K. Friedrich, D. Bhattacharyya, A direct comparison of matched-die versus diaphragm forming, *Composites Part A* 29A (1998) 183–188.
- [58] W.R. Yu, M. Zampaloni, F. Pourboghrat, K. Chung, T.J. Kang, Sheet hydro-forming of woven FRT composites: non-orthogonal constitutive equation considering shear stiffness and undulation of woven structure, *Compos. Struct.* 61 (4) (2003) 353–362.
- [59] R. Akkerman, S.P. Haanappel, Thermoplastic composites manufacturing by thermoforming, in: P. Boisse (Ed.), *Advances in Composites Manufacturing and Process Design*, Woodhead Publishing 2015, pp. 111–129.
- [60] F.O. Sonmez, M. Akbulut, Process optimization of tape placement for thermoplastic composites, *Compos. Appl. Sci. Manuf.* 38 (9) (2007) 2013–2023.
- [61] K. van Rijswijk, S. Lindstedt, D.P.N. Vlasveld, H.E.N. Bersee, A. Beukers, Reactive processing of anionic polyamide-6 for application in fiber composites: a comparative study with melt processed polyamides and nanocomposites, *Polym. Test.* 25 (7) (2006) 873–887.
- [62] I. Sibikini, J. Karger-Kocsis, Toward industrial use of anionically activated lactam polymers: past, present and future, *Adv. Indust. Eng. Polym. Res.* 1 (1) (2018) 48–60.
- [63] R.A. Witik, R. Teuscher, V. Michaud, C. Ludwig, J.-A.E. Manson, Carbon fibre reinforced composite waste: an environmental assessment of recycling, energy recovery and landfilling, *Compos. Appl. Sci. Manuf.* 49 (2013) 89–99.
- [64] D. Ayre, Technology advancing polymers and polymer composites towards sustainability: a review, *Curr. Opin. Green Sustain. Chem.* 13 (2018) 108–112.
- [65] J. Chen, J. Wang, A. Ni, Recycling and reuse of composite materials for wind turbine blades: an overview, *J. Reinforc. Plast. Compos.* 38 (12) (2019) 567–577.
- [66] G. Oliveux, L.O. Dandy, G.A. Leeke, Current status of recycling of fibre reinforced polymers: review of technologies, reuse and resulting properties, *Prog. Mater. Sci.* 72 (2015) 61–99.
- [67] K.I. Budhwani, M. Janney, U.K. Vaidya, Vibration-based nondestructive testing to determine viability of parts produced with recycled thermoplastic composites, *Mater. Eval.* 74 (2) (2016) 181–193.
- [68] K. Behloul, J. Mérotte, J. Le Bihan, N. Renouard, A. Kervoëlen, M. Fournet, A. Bourmaud, Études des voies de revalorisation pour des composites non tissés poly(propylène)/fibre de lin, *Rev. Compos. Matériaux Avancés* 26 (3–4) (2016) 295–311.
- [69] S.-Y. Fu, B. Lauke, Y.-W. Mai, *Science and Engineering of Short Fibre Reinforced Polymer Composites*, Woodhead Publishing Limited, Cambridge (UK), 2009.
- [70] B. Agarwal, L. Broutman, K. Chandrashekhara, *Analysis and Performance of Fiber Composites*, third ed., John Wiley & Sons, New York, USA, 2017.
- [71] N. Graupner, K. Albrecht, G. Ziegmann, H. Enzler, J. Muessig, Influence of reprocessing on fibre length distribution, tensile strength and impact strength of injection moulded cellulose fibre-reinforced polylactide (PLA) composites, *Express Polym. Lett.* 10 (8) (2016) 647–663.
- [72] A. Vaxman, M. Narkis, A. Siegmans, S. Kenig, in: L. Nicolais, A. Borzacchiello (Eds.), *Short-Fiber Thermoplastics Composites: Fiber Fracture during Melt Processing*, Wiley Encyclopedia of Composites, John Wiley & Sons, Hoboken, NJ, USA, 2012.
- [73] J.L. Thomason, Glass fibre sizing: a review, *Compos. Appl. Sci. Manuf.* 127 (2019).
- [74] L. Yang, J.L. Thomason, W. Zhu, The influence of thermo-oxidative degradation on the measured interface strength of glass fibre-polypropylene, *Compos. Appl. Sci. Manuf.* 42 (10) (2011) 1293–1300.
- [75] A. Inoue, K. Morita, T. Tanaka, Y. Arao, Y. Sawada, Effect of screw design on fiber breakage and dispersion in injection-molded long glass-fiber-reinforced polypropylene, *J. Compos. Mater.* 49 (1) (2013) 75–84.
- [76] Á. Kmetty, T. Bárány, J. Karger-Kocsis, Self-reinforced polymeric materials: a review, *Prog. Polym. Sci.* 35 (10) (2010) 1288–1310.
- [77] K.P. Matabola, A.R. De Vries, F.S. Moolman, A.S. Luyt, Single polymer composites: a review, *J. Mater. Sci.* 44 (23) (2009) 6213–6222.
- [78] A. Pegoretti, Trends in composite materials: the challenge of single-polymer composites, *Express Polym. Lett.* 1 (11) (2007) 710.
- [79] I.M. Ward, P.J. Hun, The science and technology of hot compaction, *Polymer* 45 (2004) 1413–1427.
- [80] J. Karger-Kocsis, T. Bárány, Single-polymer composites (SPCs): status and future trends, *Compos. Sci. Technol.* 92 (2014) 77–94.
- [81] S. Fakirov, D. Bhattacharya, *Handbook of Engineering Biopolymers: Homopolymers, Blends and Composites*, Hanser Publisher, Munich, 2007.
- [82] D. Bhattacharya, S. Fakirov, *Synthetic Polymer-Polymer Composites*, Hanser Publisher, Munich, 2012.
- [83] S. Fakirov, Nano-/microfibrillar polymer-polymer and single polymer composites: the converting instead of adding concept, *Compos. Sci. Technol.* 89 (2013) 211–225.
- [84] S. Fakirov, Nano- and microfibrillar single-polymer composites: a review, *Macromol. Mater. Eng.* 298 (1) (2013) 9–32.
- [85] R.J. Shields, D. Bhattacharyya, S. Fakirov, Fibrillar polymer-polymer composites: morphology, properties and applications, *J. Mater. Sci.* 43 (20) (2008) 6758–6770.
- [86] N.J. Capiati, R.S. Porter, The concept of one polymer composites modelled with high density polyethylene, *J. Mater. Sci.* 10 (1975) 1671–1677s.
- [87] N.-M. Barkoula, T. Peijs, T. Schimanski, J. Loos, Processing of single polymer composites using the concept of constrained fibers, *Polym. Compos.* 26 (1) (2005) 114–120.
- [88] B. Alcock, N.O. Cabrera, N.M. Barkoula, Z. Wang, T. Peijs, The effect of temperature and strain rate on the impact performance of recyclable all-polypropylene composites, *Compos. B Eng.* 39 (3) (2008) 537–547.
- [89] B. Alcock, N.O. Cabrera, N.M. Barkoula, T. Peijs, Direct forming of all-polypropylene composites products from fabrics made of Co-extruded Tapes, *Appl. Compos. Mater.* 16 (2) (2009) 117–134.
- [90] B. Alcock, N. Cabrera, N. Barkoula, T. Peijs, Low velocity impact performance of recyclable all-polypropylene composites, *Compos. Sci. Technol.* 66 (11–12) (2006) 1724–1737.
- [91] L.J. Varga, T. Bárány, Development of recyclable, lightweight polypropylene-based single polymer composites with amorphous poly-alpha-olefin matrices, *Compos. Sci. Technol.* 201 (2021).
- [92] J. Rasburn, P.J. Hine, I.M. Ward, R.H. Olley, D.C. Bassett, M.A. Kabeel, The hot compaction of polyethylene terephthalate, *J. Mater. Sci.* 30 (1995) 615–622.
- [93] P. Rojanapitayakorn, P.T. Mather, A.J. Goldberg, R.A. Weiss, Optically transparent self-reinforced poly(ethylene terephthalate) composites: molecular orientation and mechanical properties, *Polymer* 46 (3) (2005) 761–773.
- [94] M. Duhovic, D. Bhattacharyya, S. Fakirov, Nanofibrillar single polymer composites of poly(ethylene terephthalate), *Macromol. Mater. Eng.* 295 (2) (2010) 95–99.
- [95] D. Bhattacharyya, P. Maitrot, S. Fakirov, Polyamide 6 single polymer composites, *Express Polym. Lett.* 3 (8) (2009) 525–532.

- [96] K. Bocz, M. Domonkos, T. Igricz, Á. Kmetty, T. Bárány, G. Marosi, Flame retarded self-reinforced poly(lactic acid) composites of outstanding impact resistance, *Compos. Appl. Sci. Manuf.* 70 (2015) 27–34.
- [97] N.K. Kim, S. Fakirov, D. Bhattacharyya, Polymer-polymer and single polymer composites involving nanofibrillar poly(vinylidene fluoride): manufacturing and mechanical properties, *J. Macromol. Sci. Part B-Physics* 53 (7) (2014) 1168–1181.
- [98] B. J. N. Al-Thani, A. Abdulkareem, Recycling of polymer-polymer composites, in: R.K. Kumar Mishra, T. S. N. Kalarikkal (Eds.), *Micro and Nano Fibrillar Composites (MFCs and NFCs) from Polymer Blends*, Woodhead Publishing Limited 2017, pp. 263–277.
- [99] T. Bárány, A. Izer, A. Menyhárd, Reprocessability and melting behaviour of self-reinforced composites based on PP homo and copolymers, *J. Therm. Anal. Calorim.* 101 (1) (2010) 255–263.
- [100] S.R. Naqvi, H.M. Prabhakara, E.A. Bramer, W. Dierkes, R. Akkerman, G. Brem, A critical review on recycling of end-of-life carbon fibre/glass fibre reinforced composites waste using pyrolysis towards a circular economy, *Resour. Conserv. Recycl.* 136 (2018) 118–129.
- [101] V. Markovic, S. Marinkovic, A study of pyrolysis of phenolic resin reinforced with carbon fibres and oxidezed PAN fibres, *Carbon* 18 (1980) 329–335.
- [102] S.J. Pickering, R.M. Kelly, J.R. Kennerley, C.D. Rudd, N.J. Fenwick, A fluidised-bed process for the recovery of glass fibers from scrap thermoset composites, *Compos. Sci. Technol.* 60 (2000) 509–523.
- [103] A.M. Cunliffe, P.T. Williams, Characterisation of products from the recycling of glass fibre reinforced polyester waste by pyrolysis, *Fuel* 82 (18) (2003) 2223–2230.
- [104] E. Lester, S. Kingman, K.H. Wong, C. Rudd, S. Pickering, N. Hilal, Microwave heating as a means for carbon fibre recovery from polymer composites: a technical feasibility study, *Mater. Res. Bull.* 39 (10) (2004) 1549–1556.
- [105] F.A. López, O. Rodríguez, F.J. Alguacil, I. García-Díaz, T.A. Centeno, J.L. García-Fierro, C. González, Recovery of carbon fibres by the thermolysis and gasification of waste prepreg, *J. Anal. Appl. Pyrol.* 104 (2013) 675–683.
- [106] L. Giorgini, C. Leonardi, L. Mazzocchi, G. Zattini, M. Cavazzoni, I. Montanari, C. Tosi, T. Benelli, Pyrolysis of fiberglass/polyester composites: recovery and characterization of obtained products, *FME Transact.* 44 (4) (2016) 405–414.
- [107] X. Su, Y. Zhao, R. Zhang, J. Bi, Investigation on degradation of polyethylene to oils in supercritical water, *Fuel Process. Technol.* 85 (8–10) (2004) 1249–1258.
- [108] L. Meng, Y. Zhang, Y. Huang, M. Shibata, R. Yosomiya, Studies on the decomposition behavior of nylon-66 in supercritical water, *Polym. Degrad. Stabil.* 83 (3) (2004) 389–393.
- [109] H. Wang, Y. Liu, Z. Li, X. Zhang, S. Zhang, Y. Zhang, Glycolysis of poly(ethylene terephthalate) catalyzed by ionic liquids, *Eur. Polym. J.* 45 (5) (2009) 1535–1544.
- [110] H. Jie, H. Ke, Q. Wenjie, Z. Zibin, Process analysis of depolymerization polybutylene terephthalate in supercritical methanol, *Polym. Degrad. Stabil.* 91 (10) (2006) 2527–2531.
- [111] L.O. Dandy, G. Oliveux, J. Wood, M.J. Jenkins, G.A. Leeke, Accelerated degradation of Polyetheretherketone (PEEK) composite materials for recycling applications, *Polym. Degrad. Stabil.* 112 (2015) 52–62.
- [112] C. Morin, A. Loppinet-Serani, F. Cansell, C. Aymonier, Near- and supercritical solvolysis of carbon fibre reinforced polymers (CFRPs) for recycling carbon fibers as a valuable resource: state of the art, *J. Supercrit. Fluids* 66 (2012) 232–240.
- [113] C.C. Kao, O.R. Ghita, K.R. Hallam, P.J. Heard, K.E. Evans, Mechanical studies of single glass fibres recycled from hydrolysis process using sub-critical water, *Compos. Appl. Sci. Manuf.* 43 (3) (2012) 398–406.
- [114] Y. Liu, J. Liu, Z. Jiang, T. Tang, Chemical recycling of carbon fibre reinforced epoxy resin composites in subcritical water: synergistic effect of phenol and KOH on the decomposition efficiency, *Polym. Degrad. Stabil.* 97 (3) (2012) 214–220.
- [115] K.H. Yoon, A.T. Dibenedetto, S.J. Huang, Recycling of unsaturated polyester resin using propylene glycol, *Polymer* 38 (9) (1997) 2281–2285.
- [116] R. Piñero-Hernanz, J. García-Serna, C. Dodds, J. Hyde, M. Poliakov, M.J. Cocero, S. Kingman, S. Pickering, E. Lester, Chemical recycling of carbon fibre composites using alcohols under subcritical and supercritical conditions, *J. Supercrit. Fluids* 46 (1) (2008) 83–92.
- [117] Y. Liu, L. Meng, Y. Huang, J. Du, Recycling of carbon/epoxy composites, *J. Appl. Polym. Sci.* 94 (5) (2004) 1912–1916.
- [118] W. Dang, M. Kubouchi, H. Sembokuya, K. Tsuda, Chemical recycling of glass fiber reinforced epoxy resin cured with amine using nitric acid, *Polymer* 46 (6) (2005) 1905–1912.