

# Recycling concepts for short-fiber-reinforced and particle-filled thermoplastic composites: A review



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## ABSTRACT

Short-fiber-reinforced thermoplastic composites (SFRTCs) and particle-filled thermoplastic composites (PFTCs) are widely used in various industrial sectors, from automotive to electronic appliances, from building constructions to domestic equipment. Their environmental impact could be significantly reduced through both open and closed loop recycling, which allows a reduction of the usage of new resources and limit the energy consumption and CO<sub>2</sub> release associated to their production. The main challenges and opportunities in the recycling of SFRTCs and PFTCs are currently represented by i) reprocessing (also called mechanical recycling), use of recycled matrices ii) use of recycled fibers, iii) use of waste composites, iv) chemical recycling. Opportunities and current limitations in the recycling strategies and technologies for SFRTCs and PFTCs are here reviewed.

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## 1. Introduction

Short-fiber-reinforced thermoplastic composites (SFRTCs) and particle-filled thermoplastic composites (PFTCs) are widely used in several applicative fields including automotive, aerospace and aeronautics, building construction, electrical equipment, sporting goods. SFRTCs consist of thermoplastic matrices reinforced with one or more discontinuous reinforcing agents such as glass fiber (GF), carbon fiber (CF), organic fiber (mostly aramid, AF), ceramic fiber and natural fiber (NF) [1]. In a typical SFRTC, relatively short fibers of variable length are randomly distributed or imperfectly aligned in continuous thermoplastic polymer matrices. Injection molding and extrusion are the most diffuse processing methods for the production of components with SFRTCs, even if additive manufacturing (AM) methods, such as fused filament fabrication (FFF), are becoming popular also with short fiber reinforced thermoplastic composites [2]. Thermoplastic matrices are generally reinforced with relatively high amounts of short fibers, typically in the range from 20 up to 50 wt%. The main microstructural parameters determining the mechanical and physical properties of SFRTCs are the fiber orientation and the fiber length (or the aspect ratio, i.e. the length/diameter ratio). Depending on the processing conditions, fiber orientation may vary from random to nearly perfectly aligned. Skin-core morphology in injection-molded component is an example of processing-induced

fiber orientation, with fibers mostly aligned along the flow direction in the outer skin layers (the one in contact with the mold surface) and lying perpendicularly to the flow direction in the core [3]. In addition, the high shear forces generated during melt processing such as compounding with twin-screw extruders and injection molding often break down the fibers thus reducing their average length and modifying their length distribution [4]. Both orientation and fiber breakage phenomena must be taken into account in evaluating the consequences of a given recycling process on the physical and mechanical properties of SFRTCs. In fact, a reinforcing fiber can efficiently increase the modulus and strength of a composite material if enough mechanical load can be transferred from the matrix to the reinforcement. Simple micromechanical considerations indicate that the maximum fiber stress increases as the fiber length increases. The upper limit is represented by the fiber stress in a unidirectional composite reinforced with continuous fibers of equal volume fraction and subjected to the same stress. The minimum fiber length required to attain the above maximum fiber stress is generally denominated as the "load transfer length" L<sub>t</sub>. When fibers are sufficiently long and the applied stress is sufficiently high, the maximum stress in the fibers is limited by the ultimate fiber strength. A critical fiber length, L<sub>c</sub>, independent of applied stress, is defined as the minimum fiber length required to stress the fibers to their ultimate strength. Fibers that are shorter than L<sub>c</sub> will pull out of the matrix under tensile load. All the above consideration are based on the assumption of a perfect fiber/matrix bonding which is seldom the case [5]. It can be easily proven that L<sub>c</sub> depends on the fiber/matrix

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<b>Nomenclature</b>	
$\eta$	viscosity
$\lambda$	thermal conductivity
$\nu$	Poisson's ratio
$\rho$	density
$\sigma_b$	stress at break
$\sigma_y$	stress at yield
$\epsilon_b$	strain at break
$\tan\delta$	loss tangent
ABS	acrylonitrile butadiene styrene
AF	aramid fiber
AM	additive manufacturing
CF	carbon fiber
CTE	coefficient of thermal expansion
D	creep compliance
DMTA	dynamic mechanical thermal analysis
E'	tensile storage modulus
E''	tensile loss modulus
E	elastic modulus
EoL	end-of-life
EPR	ethylene-propylene rubber
G'	shear storage modulus
G''	shear loss modulus
GF	glass fiber
FFF	fused filament fabrication
FLD	fiber length distribution
HDT	heat distortion temperature
HiPP	high impact polypropylene
IS	impact strength
ISS	interfacial shear strength
K <sub>IC</sub>	critical value of the stress intensity factor (fracture toughness)
M <sub>n</sub>	number average molecular weight
M <sub>t</sub>	equilibrium moisture content
M <sub>v</sub>	viscosity average molecular weight
M <sub>w</sub>	weight average molecular weight
MAPP	maleic anhydride-polypropylene copolymer
MFI	melt flow index
NF	natural fibers
PA6	polyamide-6
PA66	polyamide-66
PBT	poly(butylene terephthalate)
PC	polycarbonate
PE	polyethylene
PEEK	polyetheretherketone
PFTC	particle-filled thermoplastic composites
PET	poly(ethylene terephthalate)
PLA	polylactic acid
POM	polyoxymethylene
PMMA	poly(methyl methacrylate)
PP	polypropylene
PPS	polyphenylene sulfide
PS	polystyrene
PVC	poly(vinyl chloride)
rCF	recycled carbon fibers
rGF	recycled glass fibers
SFRTC	short-fiber-reinforced thermoplastic composites
SMA	styrene maleic anhydride
T <sub>d</sub>	decomposition temperature
T <sub>c</sub>	crystallization temperature
T <sub>g</sub>	glass transition temperature
T <sub>m</sub>	melting temperature
TEB	tensile energy to break
VST	Vicat softening temperature
wCF	waste carbon fiber
WF	wood flour
X <sub>c</sub>	crystallinity content

shear strength [1]. Therefore, in recycling processes, a crucial point is to preserve the fibers at lengths higher than L<sub>c</sub> even when repeated re-processing steps are applied [4] and to assure that a proper fiber/matrix adhesion is achieved [6,7]. A proper design of the screws of injection molding machines can help in limiting the fiber breakage process [8].

Typically, the starting raw compounds for the production of SFRTC components are 3–4 mm long cylindrical pellets, containing randomly oriented fibers 0.2–0.4 mm in length. In order to overcome this limitation, long-fiber-reinforced thermoplastics (LFTs) have been developed in which thermoplastic polymers are reinforced with fibers of 5–25 mm or longer [9]. Another issue in the recycling of SFRTC is the degradation of the mechanical properties of both matrix materials and the reinforcing fibers. In fact, in several thermoplastic matrices, re-processing often causes a thermal degradation with a decrease of the molecular weight which ultimately lead to a decrease in the mechanical properties [10]. Also the properties of the reinforcing fibers can be negatively influenced by the high temperatures at which the composites are exposed during re-processing or matrix pyrolysis [11].

Most of the studies regarding the recycling of SFRTC have been focused on composites with thermoplastic matrices such as polyethylene (PE) [12–15], polypropylene (PP) [8,16–32] polyamide 6 (PA6) [33] and polyamide 66 (PA66) [34–37], polycarbonate [38,39], poly(ethylene terephthalate) (PET) [40–44], poly(butylene terephthalate) (PBT) [45] polyphenylene sulfide (PPS) [45,46], polylactic acid (PLA) [5,47], poly(vinyl chloride) (PVC) [48],

polyoxymethylene (POM) [49], styrene maleic anhydride (SMA) [50] and poly(ether-ether-ketone) (PEEK) [51,52].

Particle-filled thermoplastic composites (PFTCs) consist of thermoplastic matrices filled with various types of reinforcing fillers [53], such as glass spheres and flakes, calcium carbonate, talc, mica, kaolin, wollastonite, montmorillonite, feldspar, carbon black, wood flour (WF), etc ... The main reasons to add a reinforcing filler to a thermoplastic matrix are i) to reduce the cost, ii) to improve the stiffness and of the dimensional stability at low and high temperatures and the impact resistance, iii) to improve the abrasion and scratch resistance, iv) to reduce the water sorption or to modify the gas permeability. In most cases, these positive effects are also accompanied by some detrimental ones, such as a decrease in the tensile strength, elongation at break and loss of optical transparency. For as the recycling of PFTCs is concerned, among the most frequently investigated thermoplastic matrices used for their preparation the attention has been mainly focused on PP [54–58], PA6 [59], PVC [60] and PET [61] reinforced with fillers such as talc, fumed silica, clays, calcium carbonate, WF and rice hulls.

## 2. Recycling strategies for short fiber reinforced thermoplastic composites (SFRTC)

The most important recycling strategies for SFRTC includes:

- mechanical recycling (re-processing);
- use of recycled matrix materials;

- use of recycled fibers;
- use of waste composites;
- chemical recycling.

The main advantages and disadvantages of SFRTC materials prepared by these different methods are summarized in the following **Table 1**.

In the following paragraphs the approaches described in **Table 1** will be described and a careful literature review will be presented.

### 2.1. Mechanical recycling (re-processing) of SFRTC

This technique consists of crushing or shredding a component at the end-of-life (EoL) followed by a fine grinding into smaller pieces. These pieces can be ultimately used to manufacture the same (closed-loop recycling) or a different (open-loop recycling) component by using the standard manufacturing processes generally adopted for SFRTCs, such as injection molding, compression molding or extrusion. Most of the studies accessible in the scientific literature are based on the approach schematically depicted in **Fig. 1**.

Several studies can be found in the scientific literature where the effects of various repeated processing steps (re-processing), such as extrusion, injection molding and compression molding, on the microstructure and thermo-mechanical properties of SFRTCs have been investigated. Some of the most relevant studies available in the scientific literature are listed in **Table 2** with indications on the type of matrix materials (PE, PP, PC, PA6, PA66, PBT, PBS, SMA PLA, PVC, PEEK), the fiber type (GF, CF, NF) and content, the number and type of re-processing steps along with the investigated materials properties.

Re-processing always resulted in a modification of the fiber length distribution [5,20,22,34,36,37,39,50]. As reported by Arbelaez et al. [20] on PP/flax-fiber composites, an important parameter affecting the fiber length after one re-processing step (mechanical grinding and injection molding) is the fiber content. The authors also added various amounts of maleic anhydride-polypropylene copolymer (MAPP) to the PP matrix as a coupling agent. As summarized in **Fig. 2**, the average fiber length after reprocessing decreases as the fiber content in the composite increases.

Being the final fiber diameter constant at about 20 µm, a reduction of about 25% in the aspect ratio of unmodified composites and of about 48% for MAPP modified composites can be calculated when the fiber content increases from 20 to 60 wt%. At the same time, it is worthwhile to observe that the fiber length is preserved to higher values when 10 wt% of maleic anhydride-polypropylene copolymer

(MAPP) is added to the PP matrix. This effect can be explained by considering that MAPP has a molecular weight lower than the PP matrix and it can therefore act as a lubricating agent. Therefore, a reduction of shearing forces among fibers and molten matrix could occur [66].

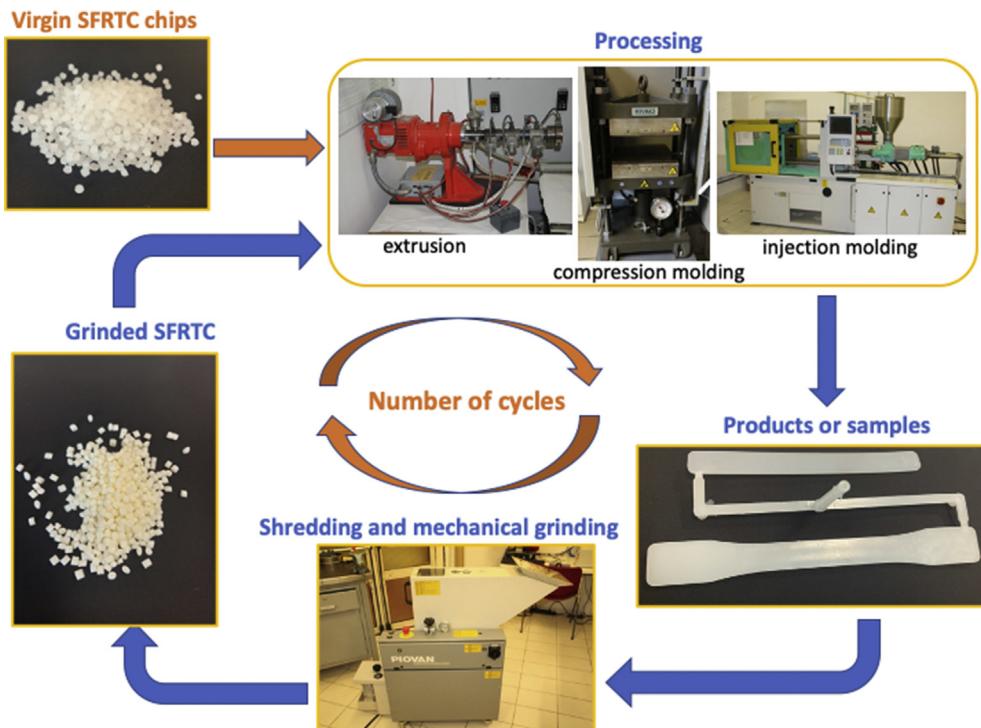
Another parameter affecting the fiber length is the number of re-processing steps the material undergoes. The influence of repeated shredding and injection molding cycles on the microstructure and mechanical properties of PP reinforced with short fibers such as GF, CF and NF (flax fibers) was recently investigated by Evans and coworkers [22]. As reported in **Fig. 3**, the length of 20 wt% of carbon fibers randomly dispersed in a polypropylene matrix dramatically decreased after repeated re-processing steps consisting of injection molding cycles. A concomitant effect was the narrowing of the fiber length distribution, also reported in **Fig. 3** as a Weibull probability plot.

From the analysis of the literature works listed in **Table 2**, it emerges that, due to the high temperature exposure and the application of elevated shear stresses, the mechanical properties of SFRTCs after re-processing may change due to the following potentially concurrent phenomena: i) shortening of the fiber length below the critical aspect ratio, ii) thermal and mechanical degradation of the polymer matrix [7]; iii) variation in the crystallinity and free volume [29,35,65] iv) variation of the fiber/matrix adhesion [7,45].

Kuram et al. [65] investigated the influence of the number of recycling steps on the mechanical, chemical, thermal and rheological properties of poly (butylene terephthalate)/polycarbonate binary blend and glass-fiber-reinforced composites. Some of the results obtained by Kuram et al. [65] are summarized in **Fig. 4**. It is interesting to observe how the tensile strength of unfilled PBT-PC blends is substantially preserved even after 5 re-processing steps. On the other hand, the tensile strength of PBT-PC/GF composites gradually decreases as the number of re-processing steps increases. This suggests that the main reason of the observed reduction in the tensile strength values of PBT-PC/GF composites can be attributed to a shortening of the fiber length and/or a reduction of the fiber/matrix adhesion rather than a degradation of the matrix. The trend of the Izod impact strength (IS) is different. In fact, the IS values measured on the neat PBT-PC matrix are initially increasing with the recycling steps but rising the number of recycling steps up to more than three times resulted in a deterioration of Izod IS values. The authors explained the observed trend by assuming that re-processing played two concurrent and counteracting effects on the PBT-PC matrix: an

**Table 1**  
Main advantages and disadvantages of most common recycling methods for SFRTC.

Recycling method	Advantages	Disadvantages
Mechanical recycling (re-processing)	<ul style="list-style-type: none"> <li>• A full recovery of both matrix and reinforcing fibers can be achieved.</li> <li>• Re-processing can be performed several times with a modest loss of properties.</li> <li>• Conventional polymer processing equipment is required.</li> </ul>	<ul style="list-style-type: none"> <li>• A decrease of fiber length is generated at each re-processing step.</li> <li>• A thermal degradation of the polymer matrix can occur at each re-processing step.</li> </ul>
Use of recycled matrix materials	<ul style="list-style-type: none"> <li>• The mechanical properties of recycled polymeric matrices can be improved by the addition of virgin or recycled fibers.</li> </ul>	<ul style="list-style-type: none"> <li>• Fiber/matrix adhesion could be an issue when non-polar matrices (such as PE and PP) are involved. Therefore, coupling agents must be added in the formulation.</li> </ul>
Use of recycled fibers	<ul style="list-style-type: none"> <li>• Short fibers coming from the recycling of continuous fiber reinforced composites can be used.</li> <li>• The cost of SFRTC composites can be decreased.</li> </ul>	<ul style="list-style-type: none"> <li>• Recycled fibers often have mechanical properties lower than the virgin ones.</li> <li>• Fiber sizing is generally removed when fibers recycled from thermal and chemical processes are used.</li> <li>• The compatibility (adhesion) between the matrix of the waste composites and the hosting matrix could be problematic.</li> </ul>
Use of waste composites	<ul style="list-style-type: none"> <li>• It represents an opportunity for the use of mechanically recycled thermosetting and thermoplastic composites after grinding.</li> </ul>	<ul style="list-style-type: none"> <li>• Hazardous solvents are often used for the fiber and/or matrix recovery from SFRTC.</li> </ul>
Chemical recycling	<ul style="list-style-type: none"> <li>• A full recovery of both matrix and reinforcing fibers can be achieved.</li> <li>• The fiber length is generally preserved.</li> </ul>	



**Fig. 1.** Schematic representation of a typical mechanical recycling (re-processing) procedure.

**Table 2**

Summary of the literature review on the mechanical recycling (re-processing) of SFRTC.

Matrix	Fiber type/content	Re-processing steps	Type of reprocessing	Measured quantities	Ref.
PE	GF/0,1,6,2,6,4,6,10 wt%	3	Extrusion	$T_m$ , $T_c$ , $h$ , $\sigma_b$ , $\varepsilon_b$	[13]
PP	NF/0,20,30,40,50,60 wt%	1, 2, 3, 4	Injection molding	FLD, E, $\sigma_b$ , $\eta$	[20]
PP	GF/0,20 wt%	1, 2, 3, 4, 5, 6, 7, 8, 9, 10	Injection molding	E, $\sigma_b$ , IS, FLD	[22]
	CF/0,20 wt%				
	NF/0,20 wt%				
PP	GF/40 wt%	1	Injection molding	E, $\sigma_b$ , $\varepsilon_b$ , $T_m$ , $T_c$ , $X_c$ , $T_d$	[29]
PP	NF/0,20 wt%	1, 3, 5	Extrusion + Injection molding	E, $\sigma_b$ , IS, $T_m$ , $T_c$ , $X_c$ , $T_d$ , $G'$ , $G''$ , $\eta$ , $T_g$ , $E'$ , $\tan\delta$	[32]
PP	GF/15,25,30 wt%	1	Injection molding	shrinkage, Shore D, $\varepsilon_b$	[62]
PP	NF/30 wt%	1, 2	Melt compounding + Compression molding	E, $\sigma_b$ , $T_d$	[63]
PC	GF/20 wt%	1, 2, 3, 4, 5	Injection molding	FLD, MFI, E, $\sigma_y$ , $\sigma_b$ , IS, $K_{IC}$	[39]
PA6	GF/28–31 vol%	1, 2, 3	Injection molding	E, $\sigma_b$	[64]
PA66	CF/30 wt%	1	Injection molding	E, $\sigma_b$ , $\varepsilon_b$ , $\eta$ , $T_g$ , $T_m$ , $T_c$ , $X_c$ , $T_d$	[35]
PA66	GF/20 wt%+CF/20 wt%	1, 2, 3, 4, 5, 6, 7, 8, 9	Injection molding	FLD, $M_w$ , $T_g$ , $T_d$ , E, $\sigma_b$ , $\varepsilon_b$ , IS	[36]
PA66	GF/0,30 wt%	1	Injection molding	FLD, IS, E, $\sigma_b$ , $\varepsilon_b$ , $E'$ , $E''$ , $\tan\delta$	[37]
PBT PPS	GF/30 wt% GF/40 wt%	1	Melt compounding + Compression molding	IS, $\sigma_b$ , $\varepsilon_b$ , $M_w$ , $X_c$ , ISS, $\tan\delta$	[45]
PBT/PC	GF/0,10 wt%	1, 2, 3, 4, 5	Injection molding	$\sigma_b$ , IS, MFI, $X_c$	[65]
SMA	GF/0,10,20,30 wt%	1, 2, 3, 4, 5	Injection molding	FLD, MFI, E, $\sigma_b$ , IS, TEB	[50]
PLA	NF/0,30 wt%	1,2,3	Injection molding	FLD, $\sigma_b$ , IS, $T_g$ , $T_m$ , $T_c$	[5]
PVC	NF/0,20 wt%	1, 2, 3, 4	Melt compounding + Compression molding	$\sigma_b$ , $\varepsilon_b$ , $\tan\delta$ , $T_d$ , $M_w$	[48]
PEEK	CF/59 vol%	1,2	Compression molding	E, $\sigma_b$	[52]

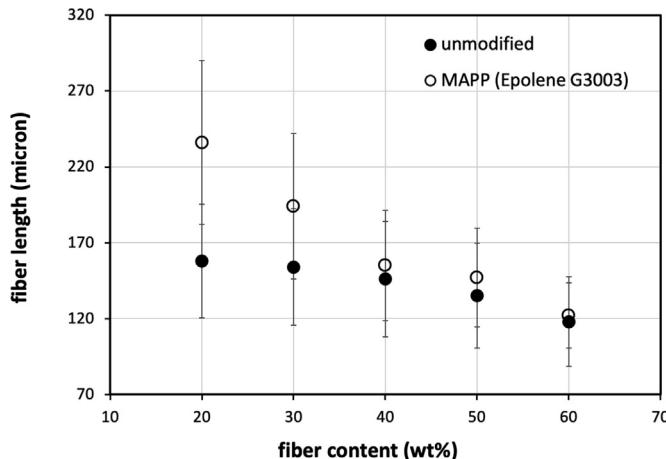
increment of its free volume (positive on the impact response) and a decrease of molecular weight (negative on the impact response). The result is a trend with a maximum value. The IS trend for the PBT-PC/GF composites is gradually increasing with the re-processing steps, most probably due to the positive effect played by the GF. Licea-Claverie et al. also observed a trend for the Charpy IS of PA66 reinforced with both 20 wt% of GF and 20 wt% of CF initially increasing up to 5 re-processing steps and then slightly decreasing [36].

The effects of reprocessing steps (granulation and injection molding) on the fracture toughness ( $K_{IC}$ ) of styrene maleic anhydride (SMA) reinforced with 10, 20 and 30 wt% of short glass fiber have been investigated by Chrysostomou and Hashemi

[50]. As reported in Fig. 5, although the standard deviation associated with each mean value is relatively high, the trend nevertheless suggests a slight decrease in fracture toughness with increasing number of re processing steps. As expected, an increase of  $K_{IC}$  with increasing the fiber volume fraction was observed.

## 2.2. SFRTC from recycled matrix materials

Another relevant strategy aimed at improving the environmental sustainability of SFRTCs is based on the usage of recycled thermoplastic matrices compounded with virgin (or sometime with recycled) fibers.



**Fig. 2.** Average fiber length after re-processing for PP/flax SFRTC as a function of the fiber content for unmodified PP and PP with 10 wt% of MAPP (data from Arbelaitz et al. [20]).

In a review paper published in 2011, Scelsi et al. [67] provided an overview on composite materials based on various different recycled thermoplastics and glass fibers. In general, they observed that the improvement of the properties that can be achieved by the addition of GF to recycled thermoplastics varied significantly system by system. Moreover, the performances of GF-filled recycled polymer composites resulted to be highly dependent on the processing methods and conditions, rather than on the specific composition of the selected polymer matrix.

Some of the more relevant studies on the use of recycled matrix materials for the preparation of SFRTC are listed in Table 3 along with some of the main parameters investigated. It can be easily noted that the majority of the research investigations have been conducted on two main types of recycled thermoplastic matrices, i.e. PP and PET. The reasons for the large interests for recycled PP are mainly related to i) its low cost, comparable or even lower that of virgin chopped GF whose price ranges from about 1.5 to 2.0 \$/kg

and ii) its large availability as end of life materials from packaging and automotive industries. On the other hand, the cost of PET is higher, but recycled PET is largely available from the collection and recycling of bottles for soft drinks and water.

Bernasconi et al. [34] investigated the tensile mechanical behaviour of PA66 injection molded composites reinforced with 35 wt% of GF and containing various percentages of mechanically recycled material (reground specimens). Tensile modulus, strength and strain at break values of PA66/GF composites with various content of reground material are summarized in Fig. 6.

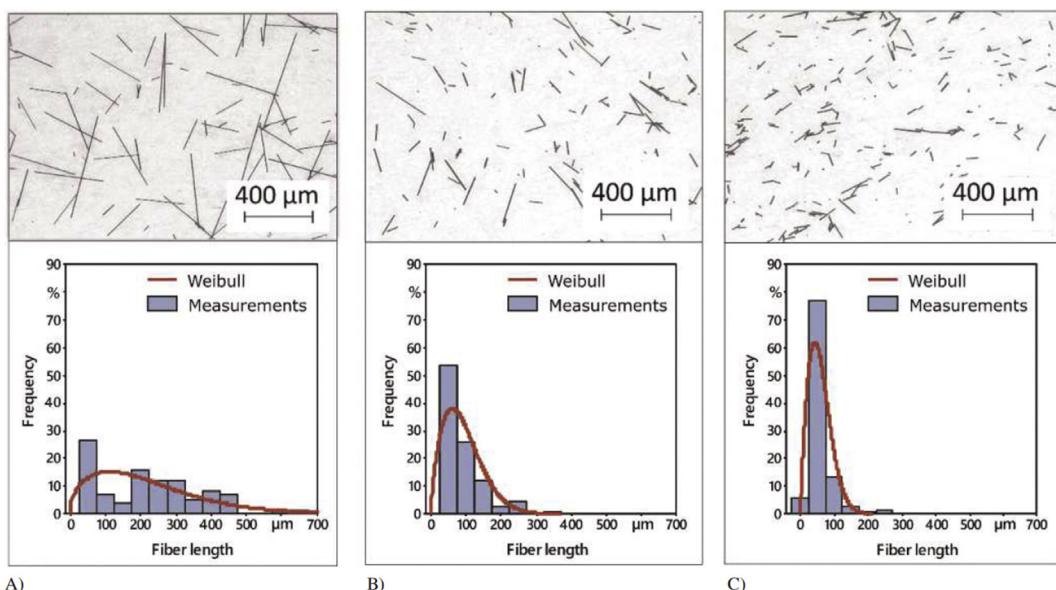
It is interesting to observe that the tensile modulus was only marginally affected by the content of reground material in the composites. On the other hand, both yield stress and strength values decreased with the content of reground material, while strain at break values were preserved or even slightly improved. These data are quite encouraging in supporting the use of recycled matrices in SFRTCs.

### 2.3. SFRTC with recycled fibers

Some efforts have been also performed to understand the possibility to use recycled fibers for the preparation of SFRTCs. Both recycled carbon fibers (rCF) and recycled glass fibers (rGF) can be obtained from thermosetting and/or thermoplastic composites by removing the polymer matrix with various methods, such as pyrolysis [72], pyro-gasification [73], solvolysis [74] and microwave thermolysis [75]. In particular, due to its high cost, most investigation have been focused on the recovery of carbon fibers from polymer composites, mostly from thermosetting matrices.

Some of the investigation available in the scientific literature on the use of recycled fibers for the preparation of SFRTCs are listed in Table 4 with some details. The recovered fibers have been used to reinforce thermoplastic matrices such as PE [72], PP [18,27,76,77], PPS [46], PLA [47], POM [49] and PA6 [78] and PA66/PA6-PE blends [79].

Burn et al. [77] investigated the usability of recycled carbon fibers in short fiber thermoplastics with a specific focus on the fiber/matrix interfacial properties. In particular, the authors simulated (pseudo-recycling) the effects of the typical recovery processes of carbon from thermosetting-based composites by



**Fig. 3.** Microscopy pictures and fiber length distribution of CF after one (A), five (B) and ten (C) injection molding cycles of PP reinforced with 20 wt% of CF. Corresponding Weibull distributions are also plotted (reprinted from Evens et al. [22], with permission of Carl Hanser Verlag GmbH & Co. KG, München).

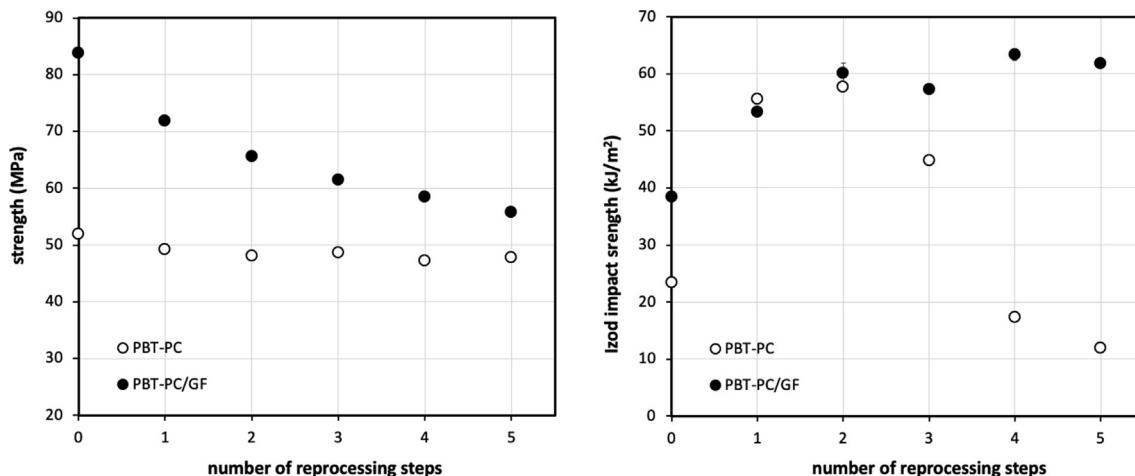


Fig. 4. Effect of re-processing steps on the tensile strength and Izod impact strength of a PBT-PC blend unfilled or reinforced with 10 wt% of GF (data from Kuram et al. [65]).

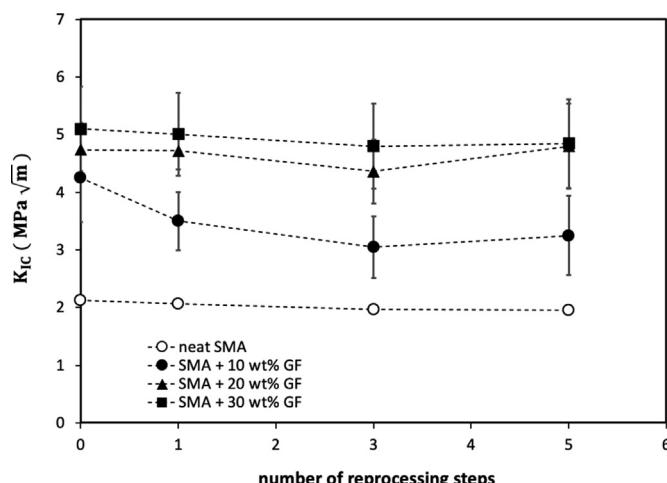


Fig. 5. Effect of re-processing steps on the fracture toughness ( $K_{IC}$ ) of styrene maleic anhydride (SMA) reinforced with 10, 20 and 30 wt% of GF (data from Chrysostomou and Hashemi [50]).

removing the sizing by pyrolysis ( $550^{\circ}\text{C}$  for 10 min in nitrogen) and also by solvolysis (washing in acetone followed by refluxing in boiling tetrahydrofuran). Both virgin and pseudo-recycled CFs were then used to prepare PP/CF single-fiber microcomposites suitable for the microdebonding test to quantify the fiber/matrix interfacial shear strength (ISS). Interestingly enough, Bum et al. [77] reported that the removal of the sizing from carbon fibers by pyrolysis and solvolysis techniques induced an alteration of the surface morphology of the fiber with a positive effect on the interfacial shear strength (IFSS) when the fibers were dispersed in a polypropylene matrix [77]. Moreover, they also proved that the reinforcing potential of rCF can be increased when the interfacial adhesion between rCF and the PP matrix was increased by the addition of 2 wt% of maleic anhydride-polypropylene copolymer (MAPP) dispersed in the PP matrix [76,77]. The ISS values obtained on PP/CF microcomposites prepared with virgin CF and with CF after pyrolysis and solvolysis embedded in neat PP and in PP with MAPP are summarized in Fig. 7. It is interesting to observe that a beneficial effect on the fiber/matrix interfacial adhesion was played by the sizing removal induced by pyrolysis and solvolysis processes. The addition of maleic anhydride had the

largest effect on the ISS values, with a 320% increase over the base polymer for the virgin fibers and 330 and 240% increase for the fibers after pyrolysis and solvolysis, respectively. Concurrently, the authors reported that the average fiber strength of the CF increased by 18–19% following sizing removal by pyrolysis and solvolysis.

#### 2.4. SFRTC from waste composites

Several investigations have proven that also waste composites could be directly used, after simply grinding in small flakes or particles, as reinforcing agents for thermoplastic matrices such as PP [31,80–83], PA6 [82], PS [84], PMMA [83], ABS [83] and PLA [85]. For example, Cerruti et al. investigated the use of a ground epoxy/CF composites coming from the tail rudder of dismantled ATR-42 aircrafts to reinforce a polystyrene matrix coming from the recycling of shipping containers [84]. They were able to homogeneously disperse up to 70 wt% of ground epoxy/CF powder into PS thus providing new all-recycled, high loading thermoplastic composites. The interesting properties shown by the all-recycled cold mixed composites suggest their use to substitute highly filled plastic components largely used, for example, in the automotive industry. Also ground unsaturated-polyester-GF and phenolic-GF composites were used to reinforce thermoplastic matrices such as PP [80–82] and PA6 [82].

Some of the information reported in manuscripts available in the scientific literature on the use of waste composites directly for the preparation of SFRTC are listed in Table 5 with some details.

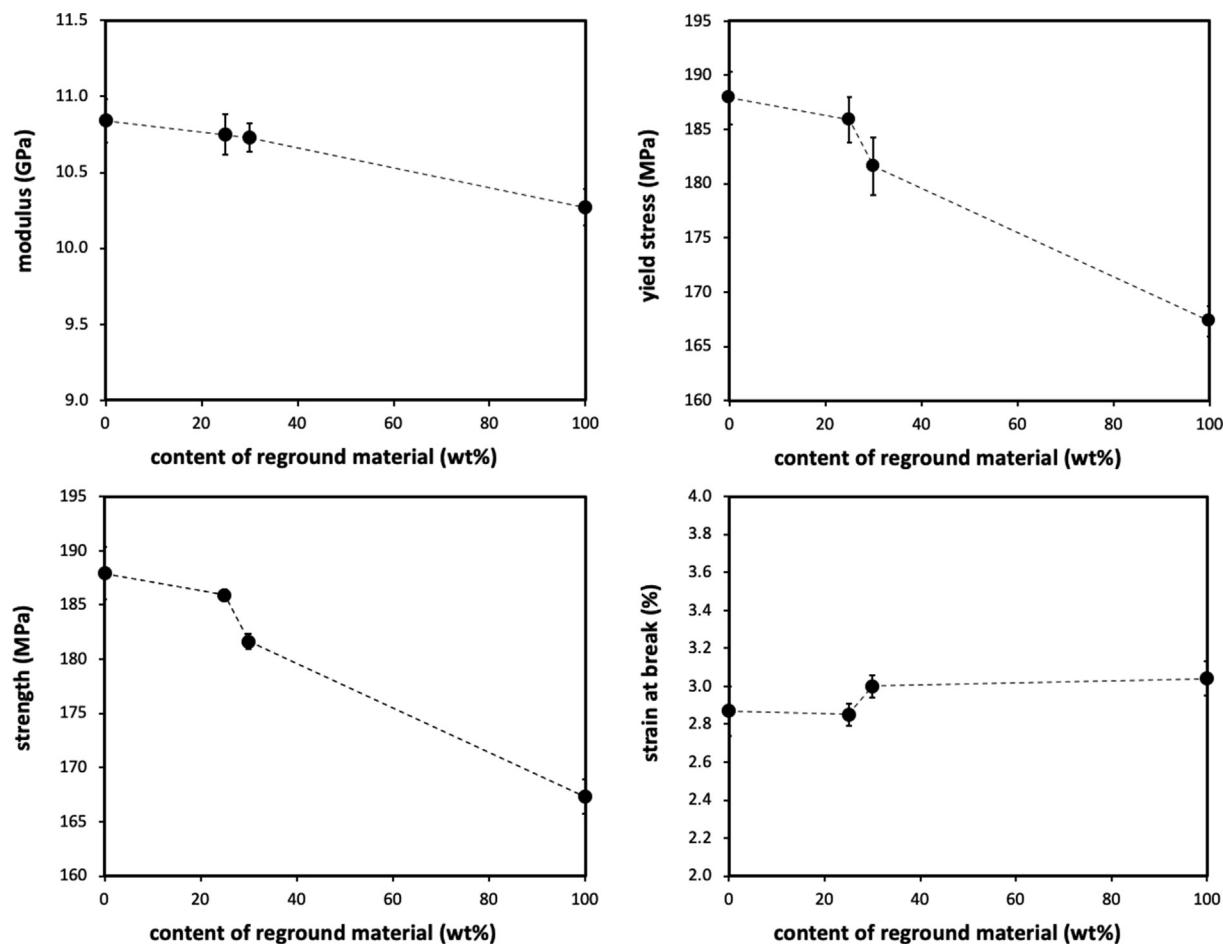
#### 2.5. Chemical recycling of SFRTC

Very limited attempts have been also reported on the recovery of short fibers from SFRTCs by chemical dissolution of the polymer matrix [51,86]. Poulakis et al. [86] investigated the possibility to dissolve PP/GF short fiber composites in a hot solvent (xylene). Subsequently, the polymer solution, containing the fibers in suspension, was filtrated for the recovery of the reinforcing agent and of the polymer matrix. The obtained constituents were successfully reused for the preparation of PP/GF short fiber composites with thermo-mechanical properties comparable to those of the virgin composites. The same concept has been also successfully proven for the recovery of carbon fibers embedded in highly stable polymer matrices such as PEEK [51]. In this case, the separation of short carbon fibers from a PEEK/CF composite required the use of drastic

**Table 3**

Summary of the literature review on the use of recycled matrix materials for the preparation of SFRTC.

Recycled matrix	Fiber type/content	Measured quantities	Ref.
PE PP	GF/0,20 wt%	$\eta$ , E, $\sigma_b$ , $\varepsilon_b$ , IS, HDT	[12]
PE	NF/0,5,10,15,20 wt%	E, $\sigma_b$ , $\varepsilon_b$ , TEB,	[14]
PE	NF/0,1,2,4 wt%	E, $\sigma_b$ , $T_m$ , $T_d$	[15]
PP	rCF/30 wt%	E, $\sigma_b$ , v, D	[21]
PP (5 and 10 reprocessing steps)	NF/0,10,30,40 wt%	$X_c$ , E, $\sigma_b$ , $\varepsilon_b$ , $K_{IC}$ ,	[25]
PP	NF/0,10,20,30,40,50 wt%	E, $\sigma_b$ , $\varepsilon_b$ , IS, $\rho$ , MFI, $T_d$ , $T_m$ , $T_c$ , $X_c$	[30]
PP	NF/0,10,20,30,40,50 wt%	$\sigma_y$ , IS, $M_t$	[68]
PP	rCF/40 wt%	FLD, E, $\sigma_b$ , D	[23]
PP	GF/30 wt%	E, $\sigma_b$ , $\varepsilon_b$ , $T_m$ , $T_c$ , $X_c$ ,	[54]
PP	rCF/25,30 wt%	Microstructure, void content	[69]
PC	GF/0,5,10,15,20 wt%	E, G', G'',	[38]
PC with impurities (ABS, PA66, PET, PC PMMA)	GF/0,5,10,15,20 wt%	$\eta$ , E, $\sigma_b$ , $\varepsilon_b$ , IS, MFI,	[70]
PET	GF/0,15,30 wt%	D	[40]
PET	GF/0,20,30,40 wt%	Mn, FLD, $X_c$ , E, $\sigma_b$ , IS	[41]
PET	GF/0,15,30 wt%	FLD, E, $\sigma_b$ , $K_{IC}$ , E', tan $\delta$ , $T_g$ , HDT,	[42]
PET	GF/0,20,30 wt%	G', h, Tg, $T_m$ , $X_c$ , E, $\sigma_b$ , $\varepsilon_b$ , IS,	[43]
PET	GF/0,15,30 wt%	$M_n$ , $T_g$ , $T_m$ , $X_c$ , $M_t$	[44]
PA66 (with 0%, 25% or 50% of virgin material)	GF/35 wt%	FLD, E, $\sigma_b$ , $\varepsilon_b$	[34]
proprietary blend of common amorphous and semicrystalline recycled polymers	GF/0,15,30 wt%	E, $\sigma_b$ , CTE	[71]

**Fig. 6.** Tensile modulus, strength and strain at break of PA66/GF composites with various content of reground material (data from Bernasconi et al. [34]).

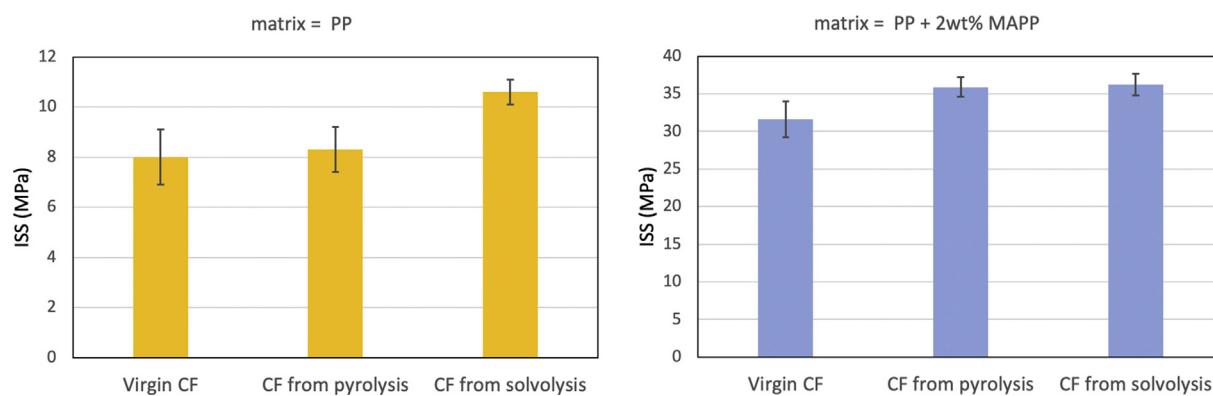
conditions by a high-pressure bomb reactor at a pressure of  $16.8 \pm 0.2$  MPa. In particular, the decomposition of PEEK/CF composites was carried out at  $350^\circ\text{C}$  by using a co-solvent system comprising of ethanol and water, incorporating catalytic quantities of caesium carbonate. The decomposition is effective within a

relatively short reaction time of 30 min, allowing for the separation of carbon fibers from the polymer matrix. Relatively clean carbon fibers were obtained without washing, however the presence of residual quantities of caesium carbonate was revealed by Energy Dispersive X-Ray spectrometry.

**Table 4**

Summary of the literature review on the use of recycled fibers for the preparation of SFRTC.

Matrix material	Recycled fiber type/content	Measured quantities	Ref.
PE	rCF/15 wt% rGF/15 wt%	E, $\sigma_b$ , $\epsilon_b$ , IS	[72]
PP	Fibers from TV panels/20 wt%	E, $\sigma_b$ , $\epsilon_b$ , IS, HDT, $T_g$	[18]
PP	rCF/50 wt% wCF/50 wt%	FLD, E, $\sigma_b$	[27]
PP	rCF/30 wt%	E, IS	[76]
PP	rCF/single fiber composites	ISS (microbond test)	[77]
PPS	rCF/20,40 wt%	E, $\sigma_b$ , $\epsilon_b$ , IS	[46]
PLA	rCF/0,10,20,30 wt%	E, $\sigma_b$ , $\epsilon_b$ , IS, $T_g$ , $T_m$ , $T_c$ , $X_c$	[47]
POM	rCF/0,5,10,15,20 wt%	E, $\sigma_b$ , $\epsilon_b$ , IS, $T_g$ , $T_m$ , $T_c$ , $X_c$	[49]
PA6	rCF/20 (including 0,1,2,5 wt% GF)	E, $\sigma_b$ , $\epsilon_b$	[78]
PA66/PA6-PE blend	rCF/20,30,40,vol%	E, $\sigma_b$ , $\epsilon_b$ , IS, fiber orientation	[79]

**Fig. 7.** Interfacial shear strength values of PP/CF microcomposites obtained with virgin CF and with CF after pyrolysis and solvolysis embedded in neat PP and in PP additivated with MAPP (data from Burn et al. [77]).

### 3. Recycling strategies for particle filled thermoplastic composites (PFTC)

#### 3.1. Mechanical recycling (re-processing) of PFTC

For as particle filled thermoplastic composites are concerned, the dominant route for recycling is re-processing according to the scheme indicated in Fig. 1. In most cases the properties of PFTC were investigated as a function of the number of recycled steps, but in other cases the effects of processing time was studied [10,87]. The most relevant studies available in the scientific literature on the mechanical recycling (re-processing) of PFTC are listed in Table 6, along with some of the information herein reported.

In a study on high-impact polypropylene (HiPP) with and without the addition of 12 wt% of talc, Bahloul et al. reported on the effects of repeated reprocessing steps (extrusion and injection molding) on the structure and thermo-mechanical properties

[56]. Some mechanical data are summarized in Fig. 8, where the tensile modulus, strength and strain at break of both HiPP and HiPP/talc are plotted as a function of consecutive processing steps. As expected, the tensile modulus improved when a filler such as talc was added to the polymer matrix. At the same time, it is worthwhile to note that the tensile modulus of neat HiPP was not substantially affected by the repeated re-processing cycles and the same trend can be observed for the HiPP/talc composites. This means that the mechanical response in the small deformation range was preserved. On the other hand, it can be observed that both stress and strain at break decreased when talc was added. This effect is well documented in particle filled systems due to the rather poor load bearing capability of a particulate filler and the difficulties in promoting a good filler/matrix adhesion level. It is also interesting to observe how both stress and strain at break values markedly decreased when repeated re-processing cycles were applied. This suggests that a reduction of the filler/

**Table 5**

Summary of the literature review on the use of waste composites directly for the preparation of SFRTC.

Matrix material	Waste composite/content	Measured quantities	Ref.
PP	Epoxy-CF/0,1,5,7 wt%	FLD, MFI, $T_m$ , $T_c$ , $X_c$ , E, $\sigma_b$ , $\epsilon_b$	[31]
PP	Unsaturated-GF/42–47 wt%	FLD, E, $\sigma_y$ , IS	[80,81]
PP	Unsaturated-polyester-GF/90 wt%	$\sigma_b$ , IS	[83]
PMMA	Unsaturated-polyester-GF/60 wt%		
ABS	Unsaturated-polyester-GF/70 wt%		
PP	Phenolic-GF/0,20 wt%	FLD, E, $\sigma_b$	[82]
PA6			
PS	Epoxy-CF/50,70 wt%	E, $\sigma_b$ , IS, $T_d$ , $\lambda$	[84]
PLA	Epoxy-GF/0,5,10,15,20,25 wt%	FLD, E, $\sigma_b$ , $\epsilon_b$	[85]

**Table 6**

Summary of the literature review on the mechanical recycling (re-processing) of PFTC.

Matrix	Filler type/content	Re-processing steps/time	Type of reprocessing	Measured quantities	Ref.
PE	Wood flour/60 wt%	1 Extrusion		MFI, water uptake, $\rho$ , E, $\sigma_b$ , $\varepsilon_b$ , $\eta$ ,	[88]
PE	Fumed silica/0.2 vol%	15,30,45,60,90,120,180,360 min Melt compounding + Compression molding		MFI, $T_m$ , $T_c$ , $X_c$ , E, $\sigma_b$ , $\varepsilon_b$	[87]
PP	Fumed silica/0.2 vol%	5,10,15,20,30,60 min Melt compounding + Compression molding		MFI, $T_m$ , $X_c$ , E, $\sigma_y$ , $\varepsilon_b$	[10]
PP	Talc/0,10,20 wt%	1,6 Extrusion + Compression molding		$M_w$ , $X_c$ , $T_g$ , E, $\sigma_y$	[58]
PP	Talc/10,15,20 wt%	1 Injection molding		shrinkage, Shore D, $\varepsilon_b$	[62]
PP	Rice hulls/30 wt%	1,2 Melt compounding + Compression molding		E, $\sigma_b$ , $T_d$	[63]
PP	Wood flour/50 wt% Rice hulls/50 wt%	1,2,4,6 Melt compounding + Compression molding		water uptake, moisture content,	[89]
HiPP	Talc/0,12 wt% + EPDM/0,20 wt%	1,3,6,9,12 Extrusion + Injection molding		$M_n$ , $M_w$ , MFI, $T_g$ , $T_m$ , E, $\sigma_y$ , $\varepsilon_b$ , $v$ , $\eta$	[56]
PA6	Talc/0,1,2,3,4,5 phr	1,2,3		E, $\sigma_b$ , $\varepsilon_b$ , IS, $T_m$ , $T_c$ , $X_c$ , $\eta$ , colour test	[59]
PVC	Wood flour/0,65 wt%	1,2,3,4		FLD, E, $\sigma_b$ , IS, water uptake, contact angle, $\tan\delta$	[60]

matrix adhesion was the main effect of re-processing on the material microstructure.

### 3.2. PFTC from recycled matrix materials

Another strategy investigated for the development of more sustainable PFTC is the usage of recycled polymers for their preparation. A summary of the information available in the literature on the use of recycled matrix materials for the preparation of PFTCs is reported in Table 7.

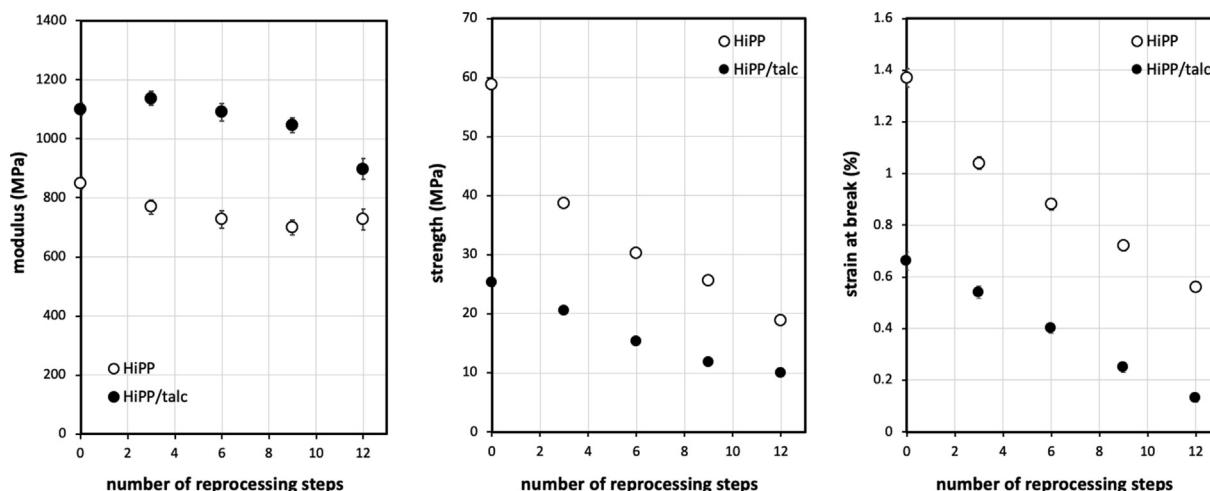
In an interesting study on PP/wood flour composites, Ares et al. [55] investigated the effects on the thermo-mechanical properties of a fraction (50%) of recycled PP mixed with the virgin polymer matrix in composites with 10, 20 and 30 wt% of wood flour (WF). As documented in Fig. 9, the tensile modulus increased while the yield stress decreased with the WF content, but both quantities were only modestly affected by the presence of 50% of recycled PP in the matrix. At the same time, it is worthwhile to note that the Vicat softening temperature (VST) increased with the WF content and

that the presence of rPP negatively affected the thermal stability of the resulting composites that displayed lower VST values.

### 4. Conclusions and future trends

According to the analysis of the existing literature, it emerges that a complete or a partial recycling of short fiber reinforced thermoplastic composites can be operated.

In particular, most of the research efforts have been focused on the full recovery of SFRTCs at the end of life by mechanical recycling consisting in grinding of the component and reprocessing. Most of the available literature studies are focused on the effects of repeated reprocessing cycles (such as injection molding or extrusion) on the microstructure and thermo-mechanical properties of SFRTCs. The most commonly reported effects of re-processing on the microstructure of SFRTCs are represented by the fiber breakage and the degradation of the molecular weight of the polymer matrix. Despite these negative effects, the mechanical properties are in most cases preserved to acceptable levels until 6–10 re-processing cycles. Other investigated possibilities are represented by i) the use

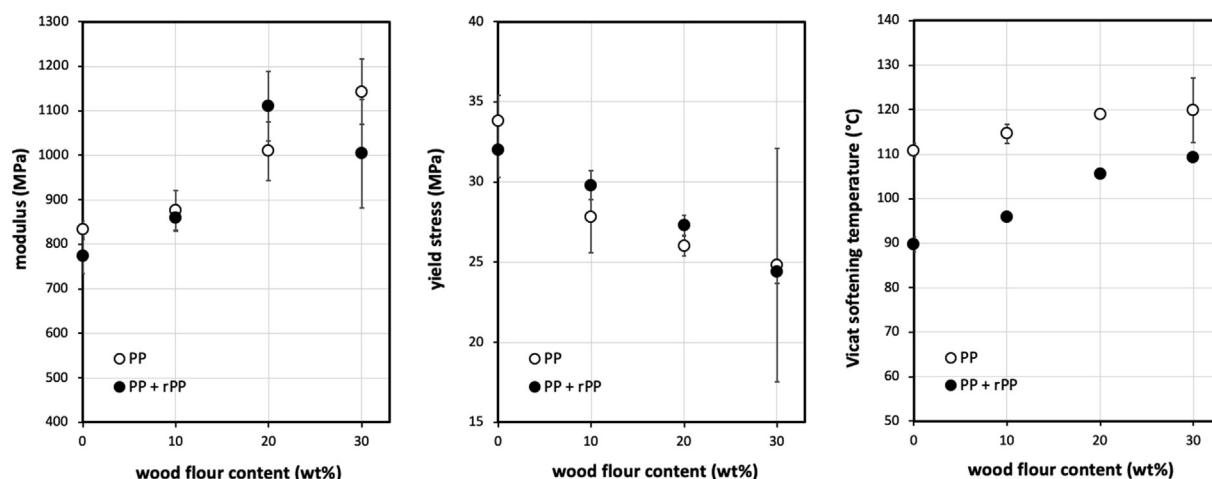


**Fig. 8.** Effect of repeated re-processing cycles (extrusion followed by injection molding) on the tensile mechanical properties of a high-impact polypropylene matrix (i.e. PP with about 20 wt% of EPDM rubber particles) unfilled (open circles) and filled without 12 wt% of talc (full circles) (data from Bahlouli et al. [56]).

**Table 7**

Summary of the literature review on the use of recycled matrix materials for the preparation of PFTC.

Recycled matrix	Fiber type/content	Measured quantities	Ref.
PP (50% blended with virgin PP)	Wood flour/0,10,20,30 wt%	E, $\sigma_y$ , $\varepsilon_y$ , $\sigma_b$ , $\varepsilon_b$ , $T_m$ , $T_c$ , $X_c$ , VST, $G'$ , $G''$ , $\eta$ ,	[55]
PP	Wood flour/32–42 wt%	E, $\sigma_b$ , $T_d$ ,	[57]
PET	Calcium carbonate/5–9 wt%		
proprietary blend of common amorphous and semicrystalline recycled polymers	Clay/1,3,5 wt%	E, $\sigma_y$ , $\varepsilon_y$ , $\sigma_b$ , $\varepsilon_b$ , D	[61]
	Calcium carbonate/20 wt%	E, $\sigma_b$ , CTE	[71]
PE	Mica/20 wt%		
	Stone wool/50 wt%	$\rho$ , E, $\sigma_b$ , IS,	[90]
	Stone dust/50 wt%		
	Primary sludge/50 wt%		
	Cellulose/50 wt%		
	Powder coating/50 wt%		
	Wood flour/50 wt%		



**Fig. 9.** Effect of 50% of recycled polypropylene (rPP) blended with virgin propylene (PP) on the tensile modulus, yield stress and VST of composites with various amounts of wood flour (data from Ares et al. [55]).

of recycled polymer matrices, ii) the use of recycled fibers, iii) the use of recycled thermosetting composites. All these strategies were proven to be effective methods to improve the thermomechanical properties of SFRTCs to levels comparable or even better than those of the virgin ones.

The possible strategies for the recycling of particle filled thermoplastic composites are mainly represented by a whole mechanical recycling (re-processing) or a partial recycling by using recycled polymer matrices. For PFTC, the main reasons for the observed decrease of thermomechanical properties due to repeated reprocessing cycles is represented by matrix degradation since filler particles are already characterized by a low aspect ratio.

Future developments in the recycling opportunities for both SFRTCs and PFTCs can be forecasted in the following directions:

- i) optimization of profiles of the screws used in injection molding and extrusion processes to reduced the fiber breakage caused by repeated re-processing steps;
- ii) re-grading of the fiber/matrix interfacial strength by the dispersion of proper coupling agents in the polymer matrices;
- iii) development of “all polymer composites” in which both matrix and reinforcing components are made with thermoplastic polymers, in some cases the polymers have the same

chemical nature and therefore the material can be melted at the end of the life for a full recovery [91–95].

#### Declaration of competing interest

None.

#### 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.

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