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**DEVELOPMENT OF A RECYCLING PROCESS OF
END-OF-LIFE THERMOPLASTICS REINFORCED
WITH RECLAIMED CARBON FIBERS AND
CHARACTERIZATION OF THE SO MANUFACTURED
RECYCLED MATERIALS**

RT/2023/6/ENEA



ITALIAN NATIONAL AGENCY FOR NEW TECHNOLOGIES,
ENERGY AND SUSTAINABLE ECONOMIC DEVELOPMENT

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Abstract

This technical Report summarizes the development and implementation of a recycling process applied on end-of-life (EoL) reclaimed carbon fibers reinforced thermoplastics. Scientific activities are carried out within the REVALUE Project and are considered of primary importance in order to comply the Directive 2000/53/EC that sets out measures to prevent and limit waste from end-of-life vehicles and their components, and ensures that where possible this is reused, recycled or recovered [1]. The proposed recycling process consists in the following steps: comminution of materials by mechanical shredding and milling followed by extrusion and compression moulding.

All the experimental activities were carried out in the laboratories of the ENEA Research Centre of Brindisi and composite materials considered for these activities were polypropylene (PP) or polyamide 6 (PA6) reinforced with carbon fibers (rCFs) reclaimed by pyrolysis of decommissioned composite materials.

In the first part of this document, the main issues encountered and the results of the activities are highlighted, together with the mass and energy balances of the overall operations, with the aim collect as more as possible information in view of potential scale-up of the process.

In the second part the attention is focused on the characterization and analysis of intermediate and final products obtained by implementing the recycling process. The main goal of the characterizations are the validation of the effectiveness of the mechanical recycling process, as well as the learning of mechanical and physical properties of recycled components in view of exploitation of the results. Physical (density, fibre length, glass-transition temperature) as well as mechanical parameters (tensile strength, elastic modulus, Charpy impact energy, storage modulus and loss modulus) are measured and hereafter reported, and discussed.

Keywords: *Recycling, composite materials, thermoplastics, carbon fibers, thermal characterization, mechanical characterization.*

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1. DEVELOPMENT OF MECHANICAL RECYCLING PROCESS

In the following an in deep description of the recycling process is illustrated.

1.1. Weathering

Firstly, with the aim to simulate the natural aging that the composite materials experiment during their lifetime, components ad hoc manufactured within the project are properly weathered by means of accelerated aging. To do this, some samples are introduced in a GHIBLI laboratory oven and heated up to 120 °C for 200 hours. After conditioning, samples are submitted to tensile test, according with standard EN ISO 527-2 *Determination of tensile properties – Part 2: Test condition for moulding and extrusion plastics*, with the scope to understand how the aging affect the mechanical properties of materials.

1.2. Recycling process

The steps of the recycling process are depicted in Figure 1.



Figure 1: Flow diagram of the proposed recycling process.

1.2.1. Mechanical Shredding and Milling

First, due to the high average size of composite materials, material is shredded down to chunks below 50 mm by means of water cooled band saw, model MEP PH261 and in order to allow the feeding in the a cutting mill. Secondly, the chunks are milled further down into fragments up to about 5 mm into a cutting mill RETSCH model SM300.

Moreover, with the aim to avoid undesired turning of materials into a viscous or rubbery state due to increase of temperature during the milling, composites were previously cooled down -20 °C in a climatic chamber. The cooling step avoided both technical and safety related problems in the milling operation, preventing any softening of materials and assuring a negligible production of dispersed fibres. At the end of the milling, materials appeared in granular form with average size of 2-3 mm (Figure 2).



Figure 2: milled samples (a) and details of the cutting mill after milling (b).

1.2.2. Extrusion

Extrusion of milled materials is performed in a Thermo Scientific™ Rheomex™ 19/25 OS extruder. Downstream the extruder, some Post-ex (post extrusion) equipment were present to produce pellets by means of “*spaghetti cutting*” method. More in detail, the molten composite is continuously discharged from a die and forms continuous filaments that, entrained by gears, are cooled in a water tank and are then cut at a low temperature by a pelletizer consisting in rotating knives (Figure 3).

When PA6 is used as thermoplastic, due to its high hygroscopicity drying of composites in PA6/rCF is necessary, with the aim to prevent bubble formation and therefore brittle moulds at the end of the recycling process. The material is dried in a vacuum oven at $T=105\text{ }^{\circ}\text{C}$ for $t=8$ hours, and after left in the vacuum oven until it cooled down to room temperature (to avoid oxygenation of the material). In preliminary activities, the drying has allowed the reduction of the residual humidity from 2.1% to 0.2%, low enough to permit the extrusion without setbacks.

In order to identify the most appropriate range of process parameters for the extrusion step (temperature, rotational speed rate of endless screw, pressure), some tests were performed. First, extrusion of matrix of composite under investigation (polypropylene or polyamide 6) was carried out. Once assured that the matrix is capable of being extruded, the extrusion of the previously milled composite is performed, by adjustment of process parameters. A system capable to prevent material bridging, due to wall friction that holds up the ends of the arch, was installed into the hopper.

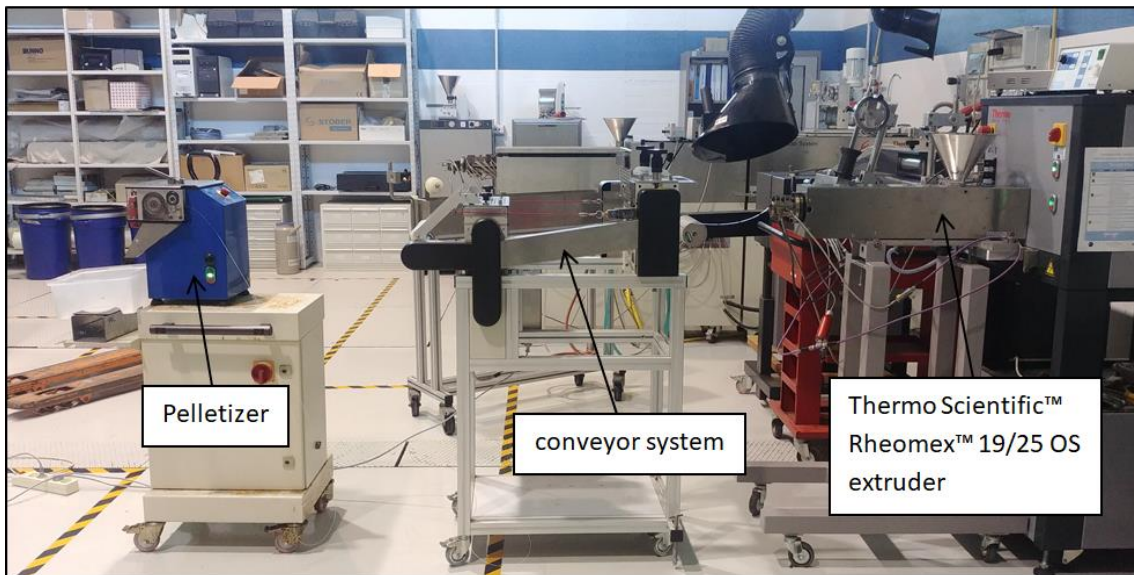


Figure 3: Extrusion and Post-ex equipment.

Outcomes of extrusion are pellets of composite material with an average length of 2.5 mm. Figure 4 depicts a detail of the continuous filament downstream the extruder and of the pellets. Details of extrusion process are reported in paragraph 1.3.

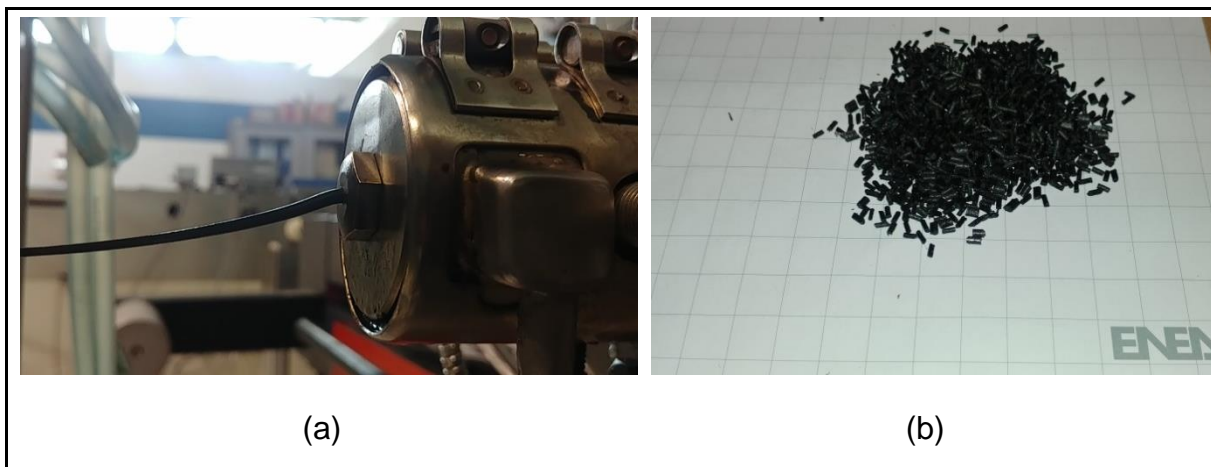


Figure 4: Detail of continuous filament formation downstream the extruder (a) and pellets (b).

1.2.3. Compression Moulding

Moulding of materials is performed in a COLLIN P 300 P/M laboratory platen press (Figure 5a), having the following parameters:

<i>Plate dimensions</i>	300 x 300 mm ²
<i>Max. force</i>	200 kN
<i>Max. temperature</i>	300 °C
<i>Heating rate</i>	up to 15 K·min ⁻¹

Cooling rate > 30 K·min⁻¹

Special features The press is equipped with a load cell which allows very precise regulation of the laminating pressure

A squared mold is used to realize the final plates: extruded material is charged into the mold (Figure 5b) and then placed inside the platen press.



Figure 5 :COLLIN P 300 P/M platen press (a) and materials charged into a squared mold (b).

The compression moulding takes place by keeping the temperature constant at 190 °C or 240 °C (if matrix is PP or PA6, respectively) and increasing the pressure from 50 to 150 bar. Moulding duration is about 18 minutes or 23 minutes (if matrix is PP or PA6, respectively), then the moulded is extracted from the platen press and sprue materials due to flashing are removed (Figures 6a and 6b). Details of compression moulding cycle and parameters are reported in section 1.3.



Figure 6: Moulded composite materials before (a) and after (b) sprue removal.

1.3. Experimental activities on preliminary composites

In order to roughly define the range of mechanical recycling process parameters, that will be further refined on composites developed and manufactured within the project (hereafter referred to as *prototypes*), some tests on preliminary materials were carried out.

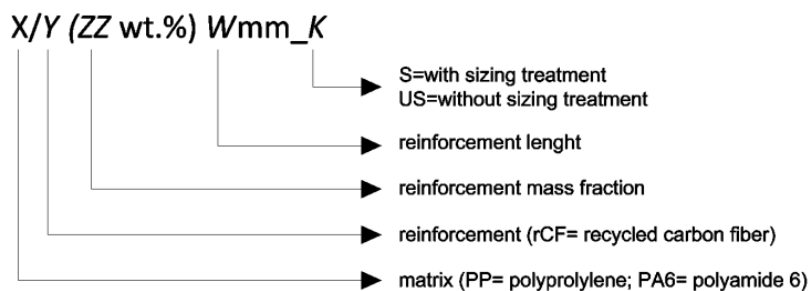
1.3.1. Materials

Six different typologies of composite materials were provided by the partner of REVALUE project CEA, in the form of dog-bone samples, sprues and heterogeneous shaped scraps and having size up to 10-15 cm. They are summarized in Table 1:

progressive number	1	2	3	4	5	6
matrix	PP	PA6	PA6	PA6	PA6	PP
reinforcement	rCFs	rCFs	rCFs	rCFs	rCFs	rCFs
mass fraction reinforcement (%)	10	10	10	30	30	30
fiber length [mm]	3	6	6	6	6	6
sized/unsized	US	US	S	US	S	US

Table 1: Preliminary composite materials that have undergone the mechanical recycling process.

The following nomenclature to refer to the composite material typologies has been introduced:



therefore the aforementioned materials will be hereinafter indicated as:

1. PP/rCF (10 wt.%) 3mm_US
2. PP/rCF (30 wt.%) 6mm_US
3. PA6/rCF (10 wt.%) 6mm_US
4. PA6/rCF (10 wt.%) 6mm_S
5. PA6/rCF (30 wt.%) 6mm_US

6. PA6/rCF (30 wt.%) 6mm_S

Figure 7 shows some samples of starting composite material subjected to the recycling process.



Figure 7: Dog-bone samples and sprues in PP/rCF (10 wt.%) 3mm_US.

Dog-bone shaped samples were used to study the accelerated weathering step, while sprues and heterogeneous shaped scraps were used to study the recycling process.

1.3.2. Preliminary tests

With the aim to simulate the natural aging that the composite materials experiment during their lifetime, a conditioning of five dog-bone shaped samples for each typology of materials was carried out, following the method described in section 1.1.

Then, the mechanical recycling process according to recipe described above has been performed on different composite materials. Process parameters for each step are listed in Table 2.

		PP/rCF	PA6/rCF
Accelerated aging	Equipment	Oven	
	Temperature	120 °C	
	Time	200 h	
Milling	Equipment	cutting mill RETSCH model SM300	
	Speed rotation	700 RPM	
Extrusion	Equipment	Thermo Scientific™ Rheomex™ 19/25 OS	
	TS1	170 °C	240 °C
	TS2	170 °C	240 °C
	TS3	170 °C	240 °C
	TS-D1	160 °C	240 °C
	n	25 RPM	20 RPM
	Equipment	COLLIN P 300 P/M laboratory platen press	

Compression Moulding	Temperature	t= 720 s @ T=190 °C	t= 720 s @ T=240 °C
		t= 400 s @ T=30 °C	t= 400 s @ T=30 °C
	Pressure	t= 300 s @ p=0 bar	t= 300 s @ p=0 bar
		p'= 1 bar/sec up to p=50 bar	p'= 1 bar/sec up to p=50 bar
		t= 200 s @ p=50 bar	t= 200 s @ p=50 bar
		p'= 1 bar/sec up to p=150 bar	p'= 1 bar/sec up to p=150 bar
		t= 480 s @ p=150 bar	t= 650 s @ p=150 bar

Table 2: Process parameters of mechanical recycling process.

In the following, data and results of experimental activities are depicted.

1.3.2.1. Mass balance

Overall recycling process was evaluated in terms of mass balance, keeping in mind that mass balance depending not only from process but also from equipment and their geometry. Results are reported in Figure 8, for all the types of materials investigated, excluded PA6/rCF (30 wt.%) 6mm_S, because of scarce starting mass processed. The yields of compression moulded materials is in the range 59.7 – 86.6% of starting weight.

As a rule of thumb, we can state that:

- shredding and milling assure a very negligible amount of dispersed fibers;
- in the extrusion process some material is lost mainly due to discarding of extruded material obtained when the process parameters were not yet optimized (too thin or too fluid filament). In permanent regime, this cause is avoided.
- A fraction of materials is lost in compression moulding, due to deflashing (removal of excess material attached to molded product).

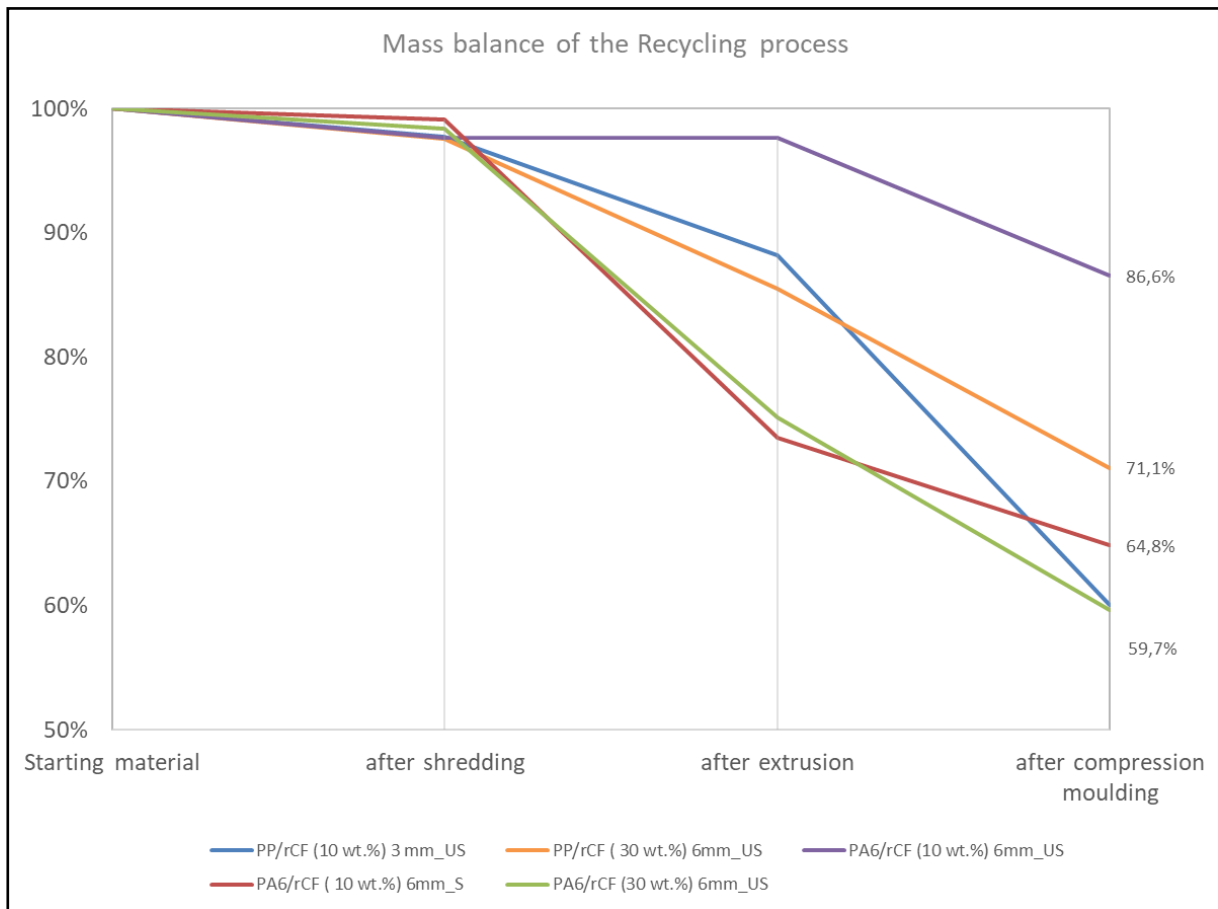


Figure 8: Mass balance of preliminary tests.

1.4. Experimental activities on manufactured prototypes

The mechanical recycling process developed on preliminary test has been replicated and, when necessary, optimized on manufactured prototypes. Outcomes of recycling process were composite plates, from which samples for mechanical characterization were obtained.

1.4.1. Materials

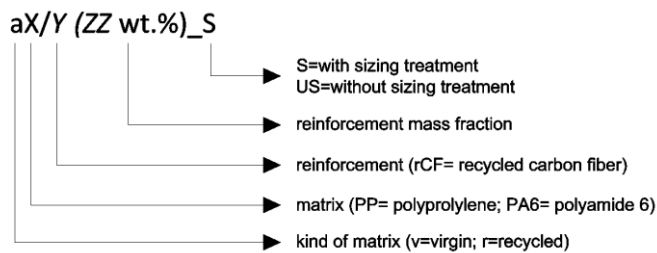
Prototypes developed and manufactured within the project were in the form of glove compartment intended to the use in the Automotive sector (Figure 9). Four different typologies of glove compartments, which differ each other in the thermoplastic polymers, were submitted to the developed recycling process: virgin polypropylene (vPP), virgin polyamide6 (vPA6), recycled polypropylene (rPP) and recycled polyamide6 (rPA6). They are listed in Table 3.

progressive number	1	2	3	4
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matrix	vPP	vPA6	rPA6	rPA6
reinforcement	rCFs	rCFs	rCFs	rCFs
mass fraction reinforcement (%)	10	10	10	10
sized/unsized	S	S	S	S

Table 3: Prototypes submitted to the mechanical recycling process.

The following nomenclature to refer to the composite material typologies has been introduced:



Therefore, the prototypes will be hereinafter indicated as:

1. vPP/rCF (10 wt.%)_S
2. vPA6/rCF (10 wt.%)_S
3. rPP/rCF (10 wt.%)_S
4. rPA6/rCF (10 wt.%)_S

In particular, in the Brindisi Research Centre ENEA laboratories the overall mechanical recycling of prototypes 1 and 2, manufactured with virgin thermoplastics, has been developed, together with the compression moulding of compounds produced by the project partner *SUEZ* by means of milling and extrusion of prototypes 3 and 4.

The following paragraphs will be therefore focused on the tests carried out on these materials.

1.4.2. Recycling process

Prototypes were provided by the partner *Centro Ricerche Fiat* (CRF) in the form of glove compartments (Figure 9). A total of 32 prototypes were provided for both types 1 and 2.

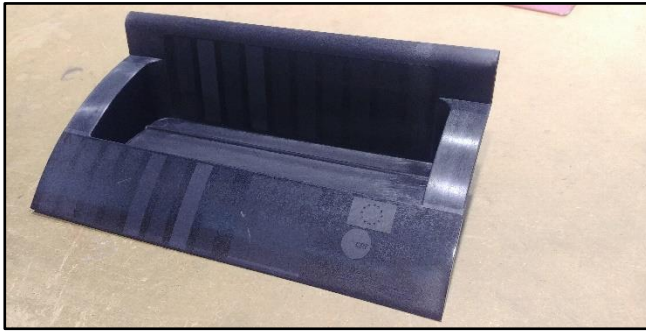


Figure 9: Glove compartment manufactured within REVALUE Project.

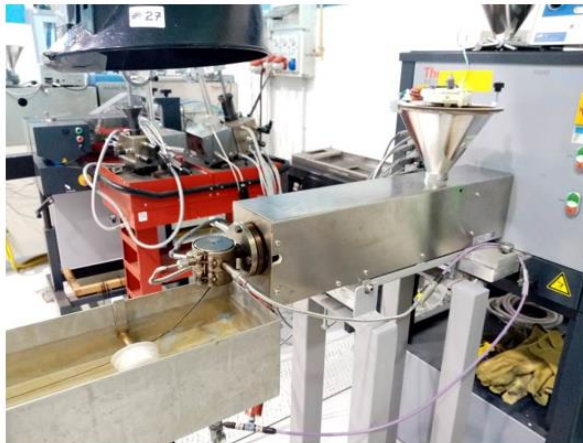
The prototypes were submitted to accelerated aging and mechanical recycling, as described in chapter 1.

Recycling process of vPA6/rCF composites went without any impediment. Unfortunately, despite of wide range of tested process parameters, the extrusion of vPP/rCF did not work and materials could not be extruded, unlike what experimented with preliminary tests described in section 1.3. The main difference between the two cases is that glove compartments were submitted to accelerated aging before the recycling process, and probably this may have caused a degradation of polymers that compromised the processability.

Considered the issues faced in the extrusion, a more in-depth analysis is shown below.

Extrusion

A picture of the lab single screw extruder, owned by ENEA Brindisi Research Centre is depicted in Figure 10. The extrusion line is equipped with a device for continuous feeding in the hopper, a cooling water tank and a pelletizer for the material granulation. A die with a 3mm hole has been used. The materials to be processed were prepared through a preliminary accelerated aging, followed by milling and subsequent sieving of the prototypes manufactured in the Project.



Technical specifications	
	Rheomex 19/25QC
Max. torque [Nm]	160
Max. speed [min^{-1}]	200
Max. Temperature [$^{\circ}\text{C}$]	450
Dimension W×H×D [mm]	230×230×700
Weight [kg]	38.5

Figure 10: Lab single screw extruder: Thermo Scientific™ Rheomex™ 19/25.

Initially, the same process parameters used in previous extrusion tests were set. In particular, for the vPA6/rCF material, no workability problems were found. The process parameters set (temperatures and screw speed) and the pellets produced are shown in Figure 11 and Figure 14.

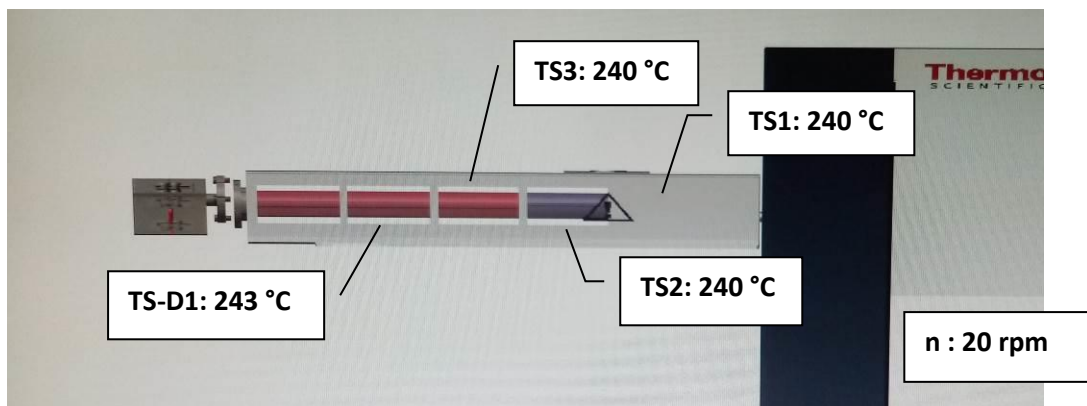


Figure 11: Process parameters for extrusion of vPA6/rCF flakes.



Figure 12: vPA6/rCF flakes



Figure 13: vPP/rCF flakes

Regarding the vPP/rCF material, the test with the initial parameters (TS1: 170°C, TS2: 170°C, TS3: 170°C, TS-D1: 160°C and n: 25 rpm) produced a discontinuous polymer melt, not suitable for pelletizing. Further tests carried out by varying the process parameters did not improve the processability of the material, which presented, in all cases, a high fluidity. This behavior is probably related to the degradation of the polymeric material, which might be due to the specific aging treatment used.



Figure 14: Extrusion process for the production of vPA6/rCF pellets.

1.4.2.1. Mass balance

A mass balance of the overall process were estimated (Table 4). Shredding, milling and extrusion were continuous process steps, therefore the mass balances are

referred to the mass flow rate. Conversely, the compression moulding step is a batch step, therefore the mass balance is referred to processed masses.

		vPP/rCF	vPA6/rCF	rPP/rCF	rPA6/rCF
Shredding & Milling	\dot{m}_{in} [g·h ⁻¹]	15000	15000	—	—
	\dot{m}_{out} [g·h ⁻¹]	14589	14892	—	—
	yield (%)	97.3	99.3	—	—
Extrusion	\dot{m}_{in} [g·h ⁻¹]	—	655	—	—
	\dot{m}_{out} [g·h ⁻¹]	—	650	—	—
	yield (%)	—	99.2	—	—
Compression Moulding	m_{in} [g]	—	475.0	410.3	475.7
	m_{out} [g]	—	426.2	333.2	421.8
	yield (%)	—	89.1	81.2	88.7
RECYCLING PROCESS	yield (%)	—	88.4	—	—

Table 4: Mass balance of recycling process.

From Table 4, it can be stated that negligible losses of materials are observed in the shredding/milling and extrusion. Conversely, a greater loss of materials occurs in the compression moulding, due to deflashing of moulded. Nevertheless, the overall yields of recycling process for vPA6/rCF (10 wt.%)_S is quite high: 88.4 %.

Moreover, it can be noticed that milling of prototypes made with vPP resulted in a little lower yield in shredding, because of a greater production of dust during the operation. Indeed, while the production of fine powder in milling of composites made with vPA6 was negligible, in case of vPP it was greater. A 1 mm sieve was used to separate milled materials from dust, that accounted for 71 wt.% and 29 wt.% of total processed composites, respectively; only the former were used for extrusion. Despite this caution, extrusion of vPP/rCF did not work, as before explained.

1.4.2.2. Energy consumption

The recycling process developed in ENEA laboratories has been evaluated also in terms of energy requirements. Specific energy consumption (*SEC*) required to carry out the steps of shredding, milling, extrusion and compression moulding have been measured and/or calculated based on process parameters adopted for the experimental activities. Because of the aforementioned difficulties in processing of vPP/rCF, hereafter only the energy consumption in recycling process of vPA6/rCF is estimated.

SHREDDING AND MILLING

No direct measurements were performed for the evaluation of energy required for shredding and milling of composites, and the specific energy consumption for these steps was therefore estimated by resorting to formulations provided in Scientific Literature. In particular, it has been estimated as a function of the mass flow rate of treated composites, as follow (2):

$$SEC_g = 11.15 \cdot \dot{m}^{-0.76} \quad [1]$$

where SEC_g is expressed in $MJ \cdot kg^{-1}$ if \dot{m} is expressed in $kg \cdot h^{-1}$.

EXTRUSION

The energy consumption during the extrusion process is due to two different contributes: the mechanical energy required to drive the material, and the thermal energy required to melt and heat the material up to the process temperature.

Driving

During the extrusion, the torque of the screw and its rotational speed are measured, from which the requested power and specific energy are determined by means of equations:

$$P = T \cdot \omega = T \cdot \frac{2\pi}{60} n \quad [2]$$

$$SEC_d = \frac{P}{\dot{m}} \quad [3]$$

Where they are:

T	Torque [N·m]
ω	angular speed [rad·s ⁻¹]
n	rotational speed [s ⁻¹]
P	power [W]
\dot{m}	mass flow rate [kg·s ⁻¹]

Heating

The thermal energy required for the heating of materials, is determined based on heater powers measured during the trials. The extruder includes three different 1 kW heaters along the screw. Another heater (0.25 kW) is present at the die section. The

overall thermal energy is the sum of each heater contribute, from which the SEC is determined as follows:

$$SEC_h = \frac{P}{\dot{m}}$$

Zone	Heater	Max power [kW]	Actual power (%)	Actual Power [kW]	SEC _h [MJ·kg ⁻¹]
Section 1	Y ₁	1.00	21.8	0.22	1.25
Section 2	Y ₂	1.00	6.8	0.07	0.39
Section 3	Y ₃	1.00	10.9	0.11	0.62
Die	Y _{D1}	0.25	35.4	0.09	0.51
Overall heating				0.48	2.76

Table 5: Thermal energy demand in extrusion.

The overall energy required for the extrusion of the material (SEC_e) is the sum of the driving and thermal contributes, which gives a value of 3.37 MJ·kg⁻¹.

COMPRESSION MOULDING

To manufacture CFRP via compression moulding, energy is required to mold the compound (*thermal energy*), to compress the material up to the required pressure (*kinetic energy*) and for the subsequent steps of cooling and finishing

Thermal energy is calculated based on the temperature profile, the specific heat (c_p) and the latent heat of fusion of polymer (L_f):

$$SEC_t = c_p \cdot \Delta T + L_f \quad [4]$$

Kinetic energy is determined based on pressing speed (v), pressure ramp rate (\dot{p}) and ram area (A_0), using the equation:

$$SEC_k = (pA - p_a A_0) v \frac{p}{\dot{p}} \quad [5]$$

The energy required for cooling and finishing are estimated from literature data and assumed equal to 0.9 MJ·kg⁻¹ and 1.2 MJ·kg⁻¹, respectively.

The overall energy required for pressing by compression moulding (SEC_{cm}) is the sum of the above-mentioned terms, which give a value of 11.98 MJ·kg⁻¹.

Using equations [1] to [5], the SEC for the overall mechanical recycling process (SEC_{rec}) has been determined (Table 6)

		vPA6/rCF
Shredding & Milling	SEC_g [MJ·kg ⁻¹]	1.42
Extrusion	SEC_e [MJ·kg ⁻¹]	3.37
Compression moulding	SEC_{cm} [MJ·kg ⁻¹]	11.98
RECYCLING PROCESS	SEC_{rec} [MJ·kg ⁻¹]	16.77

Table 6: Specific energy consumption of recycling process.

2. CHARACTERIZATION AND ANALYSIS

Analysis and characterization of both the preliminary samples and prototypes manufactured within the project were carried out to understand at what extent the mechanical recycling process affects their mechanical properties. In the following paragraphs the methods for the measurement of physical, thermal and mechanical parameters are described.

2.1. Density measurement

The density of composites was measured as the ratio between the mass of samples and their volume. Mass was measured by means of a high precision lab scale, volume was determined by immersion in a cylinder filled with water or, in case of PA6 matrix, with acetone and measuring the displaced volume of liquid.

2.2. Fiber length measurement

The average length of carbon fibers constituting the reinforcement of composites was measured, as follow:

- i. First, calcination of about 500 – 800 mg of samples of composite material was performed, in order to remove the thermoplastic polymer. Calcination was carried out in an oven at $T=450$ °C for 50 minutes.
- ii. Solid residuals of calcination, mainly carbon fibers, were observed with a light microscope and the length of fibers was estimated using the image-processing program *ImageJ*.

2.3. Tensile test

Tensile test is performed on dog-bone shaped samples of composites, to measure the ability of materials to overcome forces pulling the sample apart and the extent it stretches before breaking. The tensile test is performed according to standard ISO 527:2:2012 - *Plastics -- Determination of tensile properties -- Part 2: Test conditions for moulding and extrusion plastics*, which specifies the test conditions for determining the tensile properties of moulding and extrusion plastics, based upon the general principles given in ISO 527-1.

From the stress-strain curve obtained during the test, the following tensile properties are determined:

- **Young's modulus:** initial slope of the stress-strain curves. It is indicated as E and dimensions are GPa.
- **Yield stress:** first point on the stress-strain curves at which an increase in strain occurs without an increase in stress. It is indicated as σ_y and dimensions are MPa.
- **Tensile strength:** It describes the stress to break the sample. It also mentions the maximum tensile stress supported by specimen during test. It is indicated as σ_f and dimensions are MPa.
- **Tensile stress:** tensile load/unit area of minimum original cross section. It is expressed in force per unit area. It indicates the relationship between stress and strain in the deformation of a solid body.
- **Elongation:** increase in length produced by a tensile load. It is expressed in units of length, commonly as percentage.
- **Elongation at break:** elongation at the moment of rupture of the sample. It is indicated as ϵ and expressed as percentage.
- **Strain:** ratio of elongation to the gage length of the sample; that is the change in length per unit of original length.

2.4. Impact test

Impact test is performed on standard bars 80×10×4 mm, according to ISO 179:1:2010 *Plastics -- Determination of Charpy impact properties -- Part 1: Non-instrumented impact test*.

In the Charpy test, the specimen is supported as a horizontal beam and is broken by a single swing of a pendulum, the line of impact being midway between the supports. The test measures the **impact strength**, that is the energy removed from the pendulum as a result of the work done in breaking the test piece divided by the cross sectional area of the test piece in the direction of the swing. It provides information about the brittleness of the material.

2.5. Dynamic Mechanical Analysis (DMA)

Dynamic Mechanical Analysis (DMA) is a widely used technique to determine the temperature dependencies of the modulus and damping properties of materials by applying sinusoidal force.

Modulus is divided into two parts, **storage modulus** (E') and **loss modulus** (E''). The storage modulus represents the stiffness of a viscoelastic material, which is proportional to elastic energy stored, and is reversible. On the other hand, the loss modulus which measures the energy transferred as heat is irreversible.

E'' indicates the viscous nature of the polymer. The ratio of the loss to the storage modulus which is called **damping factor** ($\tan \delta$) is a measurement of the energy loss and represents the internal friction in a viscoelastic system.

DMA measurements were carried out using a Q800 analyzer at a heating rate of $3^{\circ}\text{C}\cdot\text{min}^{-1}$ with the frequency of 1 Hz. DMA specimens (cut from dog-bone shaped samples and others from compression moulding plates) were loaded on to the machine using a single cantilever sample holder, setting the amplitude $10\ \mu\text{m}$. The temperature was ranged from room temperature to 120°C for PA6 composites and from -30°C to 110°C for PP composites. The low starting temperature was necessary to identify the phenomenon of the glass transition, which occurs in polypropylene below the room temperature as known. E' , E'' and $\tan \delta$ were determined as a function of temperature. The **glass transition temperature** (T_g) of composites was taken by the maximum of loss modulus. Each data point was the average from two to five measurements, depending on the number of specimens available.

3. RESULTS OF CHARACTERIZATION OF PRELIMINARY SAMPLES

Analysis and characterization of the materials obtained by implementing the developed recycling process on preliminary samples, as described in paragraph 1.3, were carried out to understand at what extent the mechanical recycling process affects their mechanical properties.

3.1. Density measurement

The results of density measurements are reported in Table 7 and compared with theoretical values calculated starting from the densities of PP and PA6 (as reported on data sheets you can find in Attachment 1), rCF, and on reinforcement mass fractions. In the brackets, deviation of measured densities from theoretical ones are reported. With exception for PA6/rCF (10 wt.%) 6mm_US composites, for all the other cases the experimental density is lower than the theoretical one, suggesting the presence of voids and defects of specimens.

	Density [kg·dm ⁻³]	
	theoretical	experimental
PP/rCF (10 wt.%) 3mm_US	0.952	0.945 (-0.8%)
PP/rCF (30 wt.%) 6mm_US	1.064	1.041 (-2.2%)
PA6/rCF (10 wt.%) 6mm_US	1.183	1.199 (+1.3%)
PA6/rCF (10 wt.%) 6mm_S	1.183	1.174 (-0.8%)
PA6/rCF (30 wt.%) 6mm_US	1.281	1.090 (-14.9%)
PA6/rCF (30 wt.%) 6mm_S	1.281	1.128 (-11.9%)

Table 7: Density of samples.

3.2. Tensile test

Figures 15 to 18 depict the results of the tensile test performed on the preliminary samples after the accelerated aging test, and compared with the same parameters measured before the aging.

Excepts for PA6/rCFs (30 wt.%) 6mm _S, the Young's module does not seem to be altered from the accelerated aging, suggesting that the composites retain the elastic properties after their lifetime (Figure 15).

The results in terms of tensile strength and yield stress appear controversial. For composites made with PP, the values look unchanged after the aging if the CF

reinforcement fraction is 10 wt.%, but conversely they are strongly reduced if the CF reinforcement fraction is 30 wt.%. For composites made with PA6, after the aging it was noticed a slight increase in both the tensile strength and yield stress for composites with a CF reinforcement of 10 wt.%, and a slight decrease for those having a CF reinforcement of 30 wt.%. An exception is the composite in PA6/rCF (30 wt.%) 6mm_US, whose high standard deviation value makes the measurement unreliable (Figures 16 and 17).

The results in terms of strain at break are more consistent. Indeed, for all the investigated composites there is an undoubted reduction of the elongation at break of the specimens after aging, due to the embrittlement of materials (Figure 18).

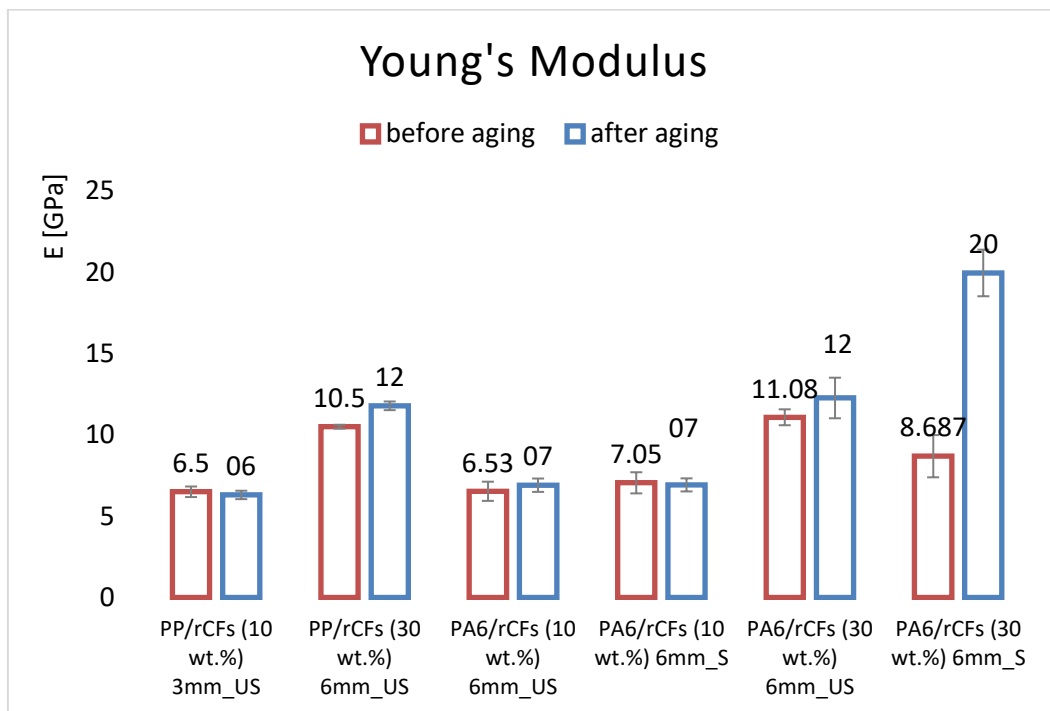


Figure 15: Young's module of composites before (red) and after (blue) the accelerated aging.

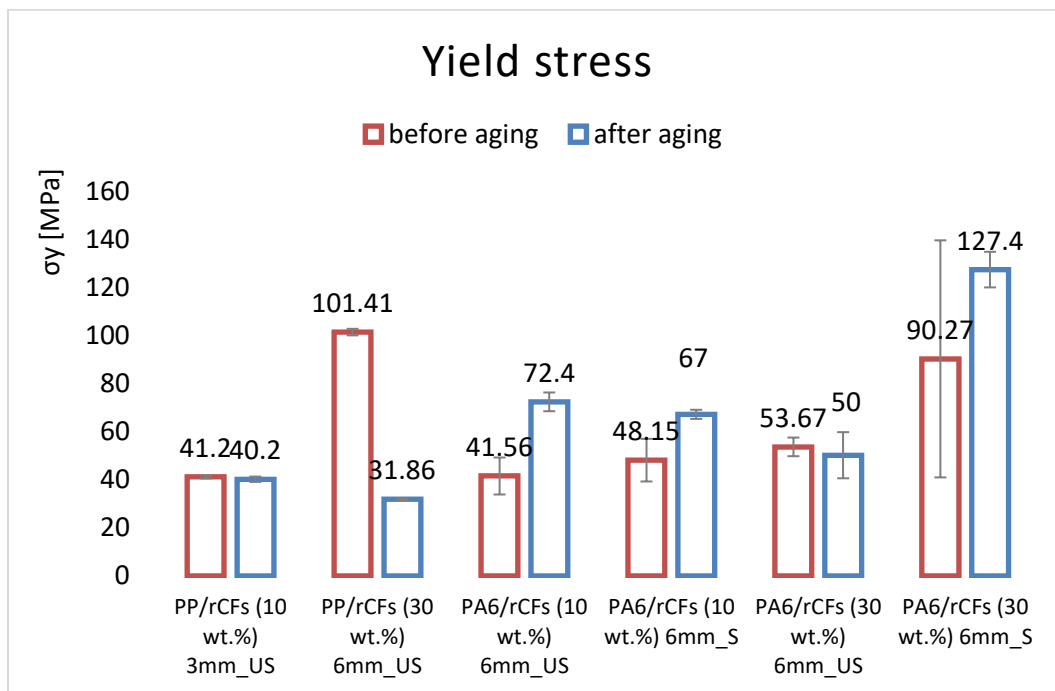


Figure 16: Yield stress of composites before (red) and after (blue) the accelerated aging.

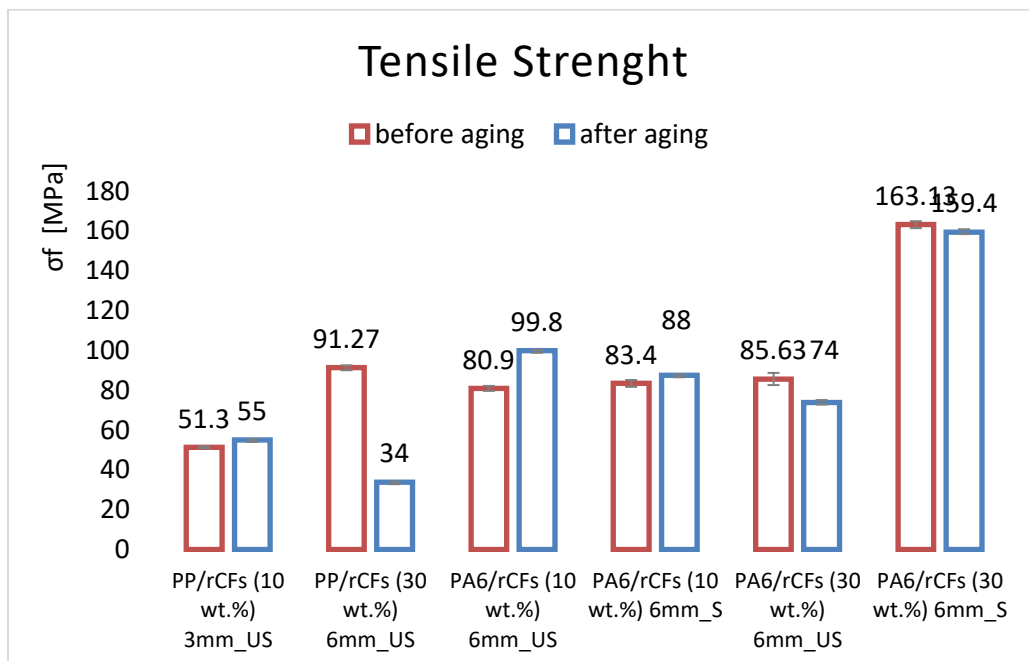


Figure 17: Tensile strength of composites before (red) and after (blue) the accelerated aging.

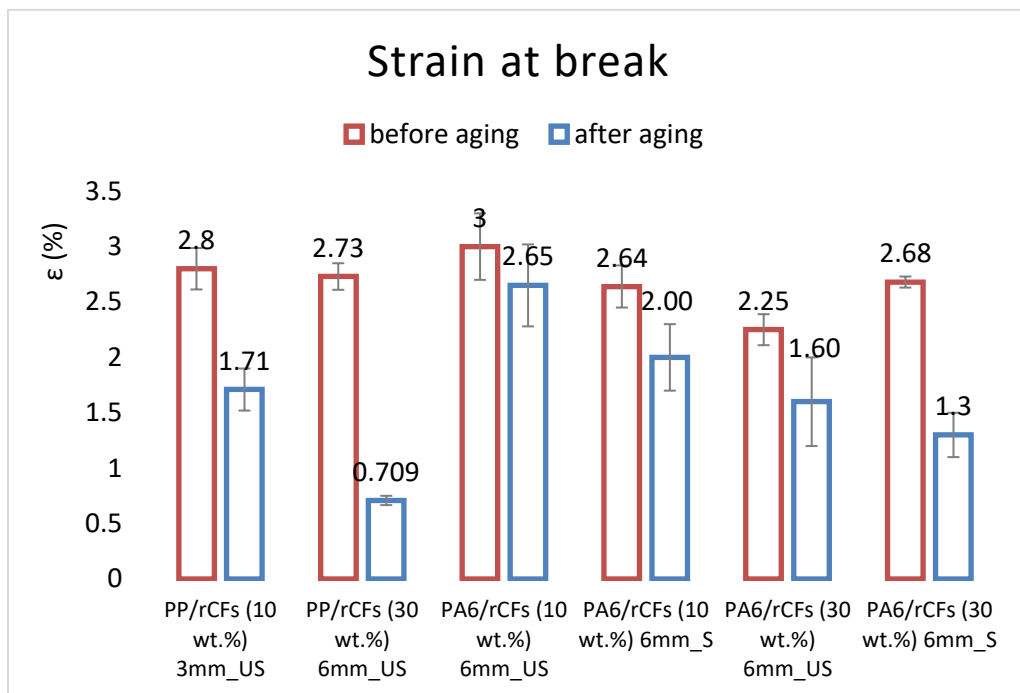


Figure 18: Strain at break of composites before (red) and after (blue) the accelerated aging.

3.3. Dynamic Mechanical Analysis

DMA tests were conducted on the preliminary PA6/rCF and PP/rCF composites samples before and after the accelerated aging test. The results of DMA tests carried out on both sized and unsized composites were reported in Figures 19 to 30 for PA6/rCF (10 wt.%) 6mm_S, PA6/rCF (10 wt.%) 6mm_US and PP/rCF (10 wt.%) 3mm_US, respectively.

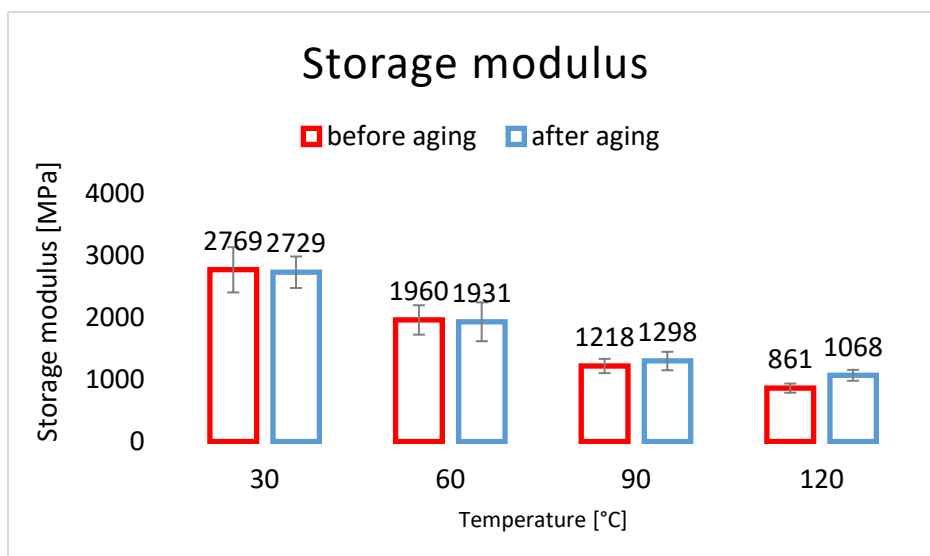


Figure 19: Storage modulus of PA6/rCF (10 wt.%) 6mm_S composites before (red) and after (blue) the accelerated aging.

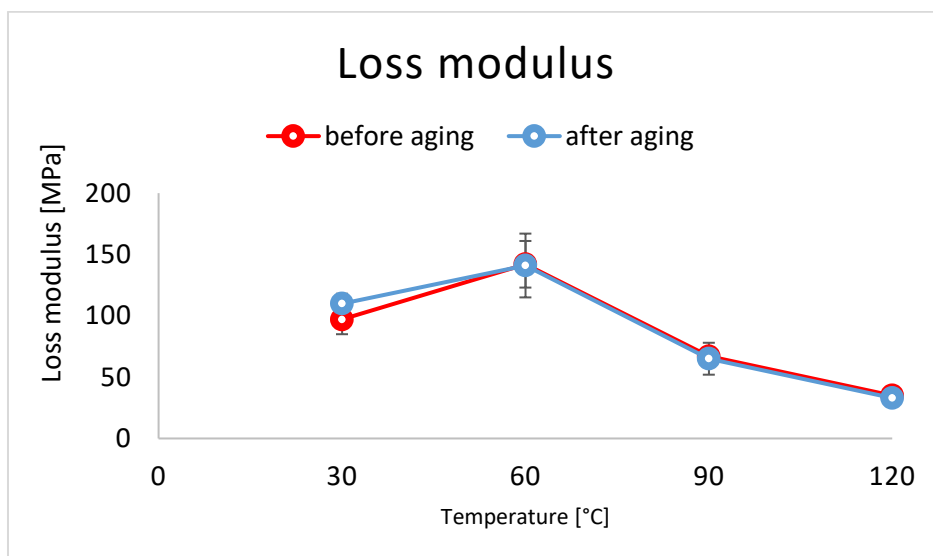


Figure 20: Loss modulus of PA6/rCF (10 wt.%) 6mm_S composites before (red) and after (blue) the accelerated aging.

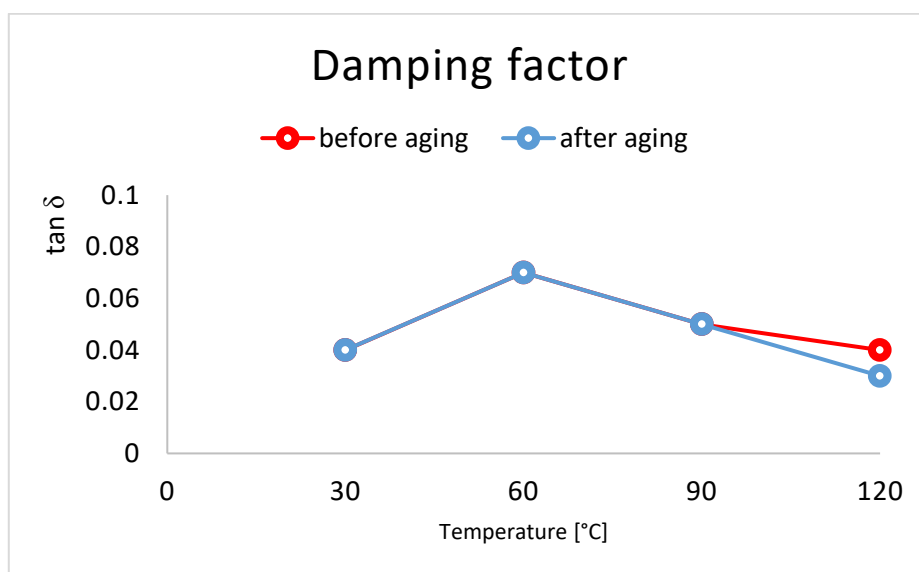


Figure 21: Tan δ of PA6/rCF (10 wt.%) 6mm_S composites before (red) and after (blue) the accelerated aging.

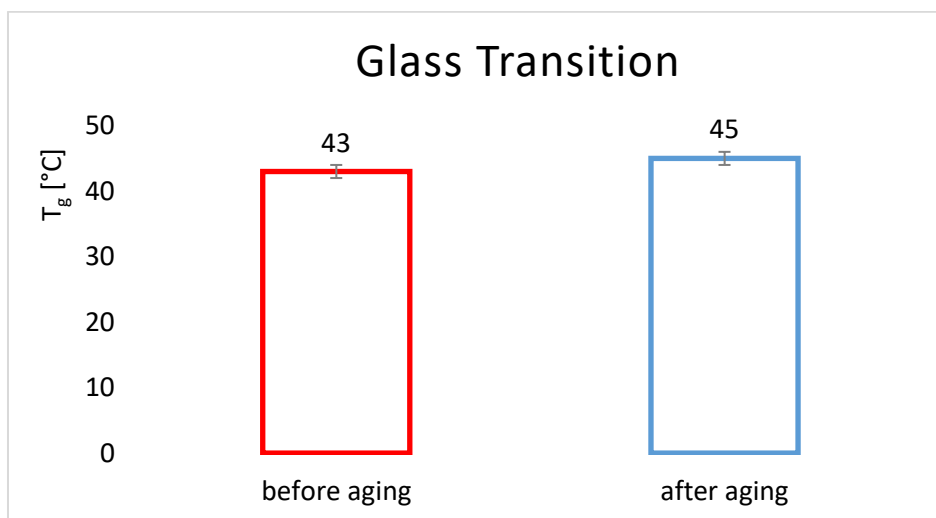


Figure 22: Glass transition temperature of PA6/rCF (10 wt.%) 6mm_S composites before (red) and after (blue) the accelerated aging.

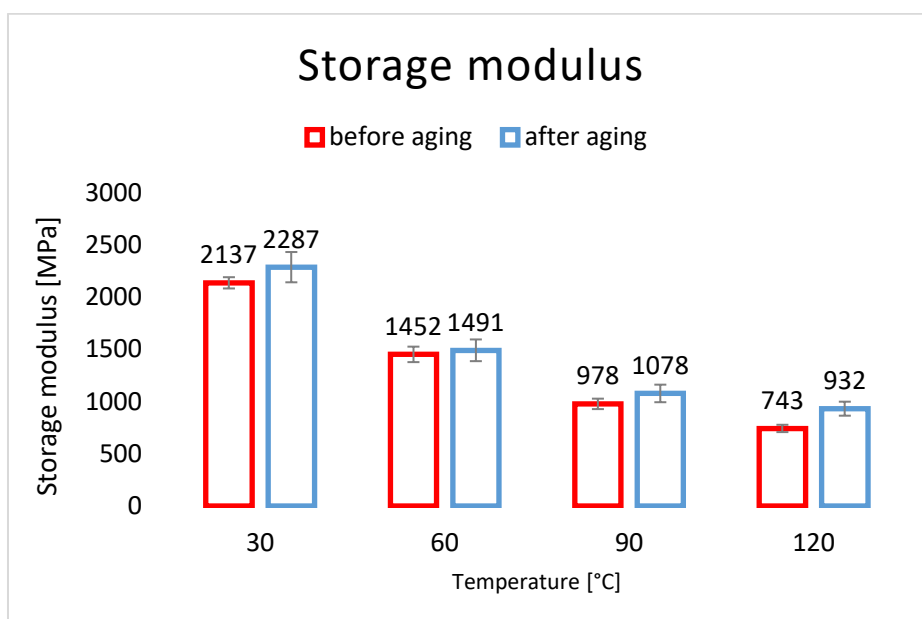


Figure 23: Storage modulus of PA6/rCF (10 wt.%) 6mm_US composites before (red) and after (blue) the accelerated aging.

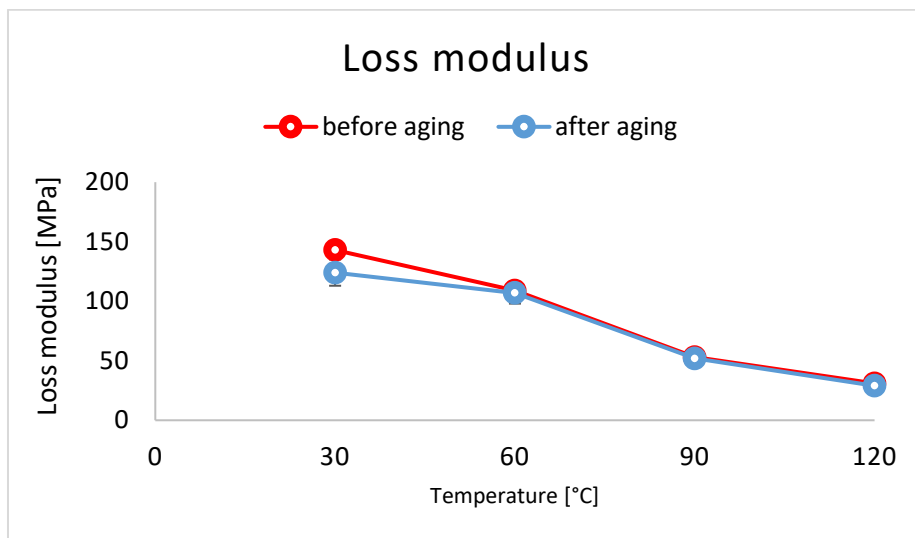


Figure 24: Loss modulus of PA6/rCF (10 wt.%) 6mm_US composites before (red) and after (blue) the accelerated aging.

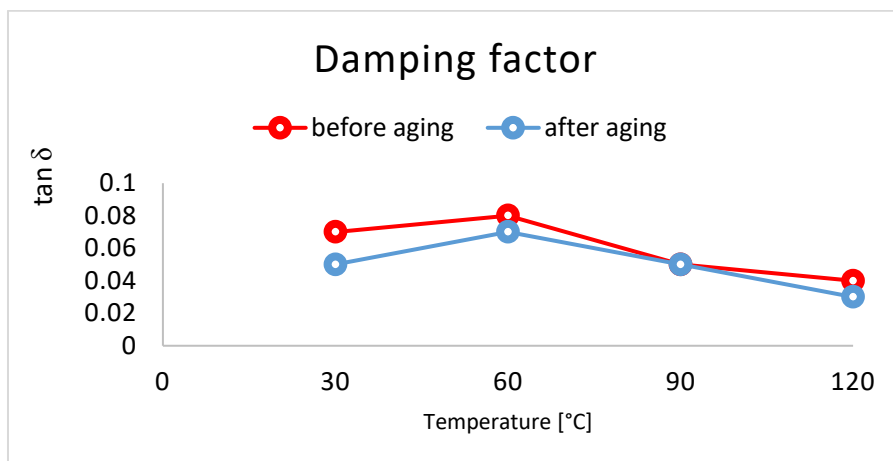


Figure 25: Tan δ of PA6/rCF (10 wt.%) 6mm_US composites before (red) and after (blue) the accelerated aging.

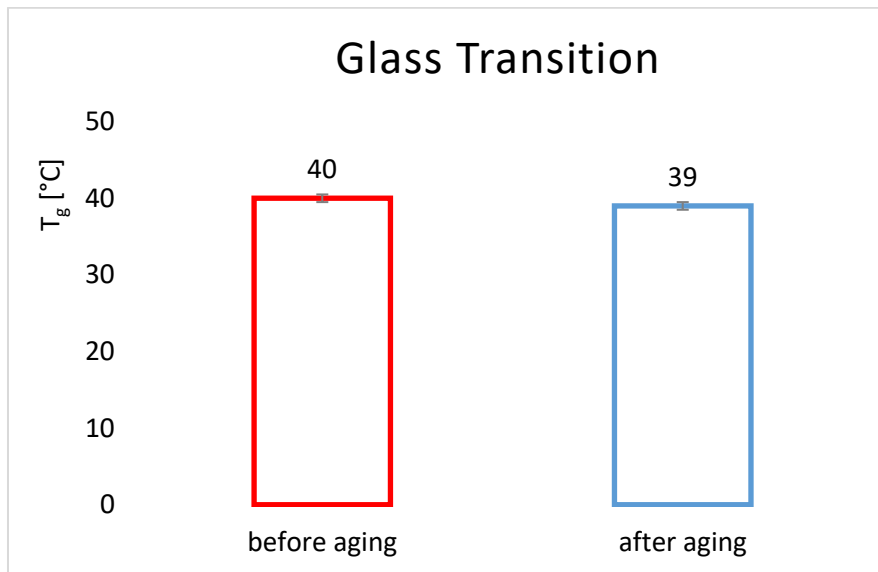


Figure 26: Glass transition temperature of PA6/rCF (10 wt.%) 6mm_US composites before (red) and after (blue) the accelerated aging.

The storage modulus was found to decrease with increase in temperature as presented in Figure 19 and in Figure 23 due to loss in stiffness on Polyamide 6 at high temperature. In addition, the aging process did not significantly affect the stiffness of the sized and unsized PA6/rCF composites material in the entire temperature range explored.

Loss modulus represented the viscous response of composites which depends upon motion of polymeric molecules in the composites. The loss modulus of composites decreased with increase in temperature (Figure 20 and Figure 24) because, as the temperature increased, the molecular mobility of polymer increased. The aging seemed not to influence the viscous nature of sized and unsized PA6/rCF composites. Moreover also the damping factor and T_g values were not modified by the accelerated aging process.

The storage modulus of aged unsized PP/rCF composites was found mildly higher than no aged materials in entire temperature range explored (Figure 27). The accelerated aging reduced the mobility of the composites by increasing the loss modulus of unsized PP/rCF compounds (Figure 28). Contrarily, no effects of aging on the damping factor were found. (Figure 29).

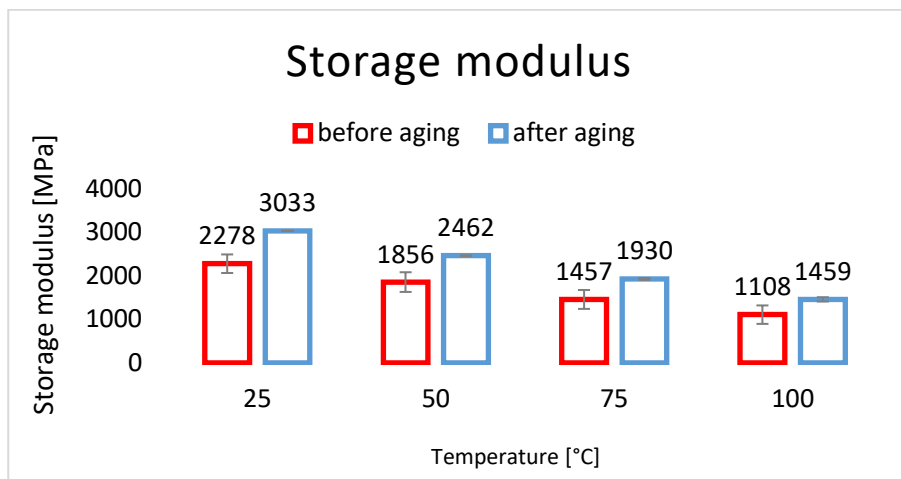


Figure 27: Storage modulus of PP/rCF (10 wt.%) 3mm_US composites before (red) and after (blue) the accelerated aging.

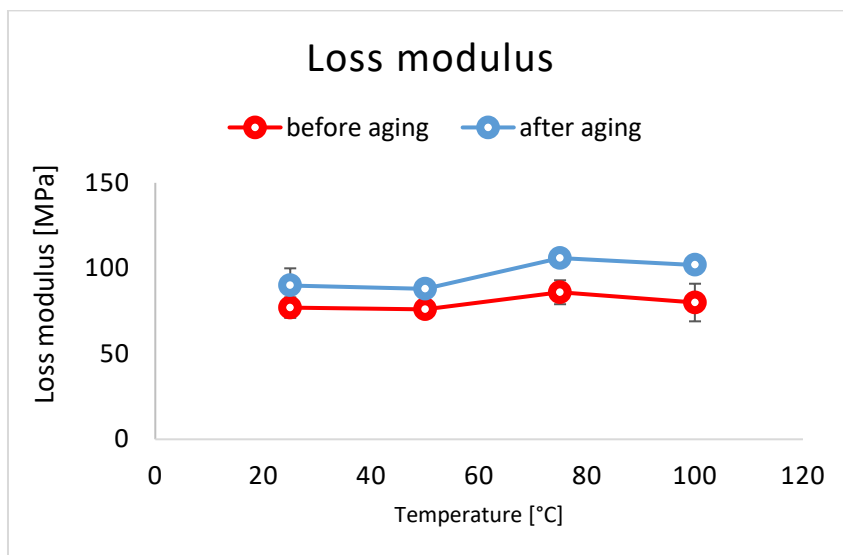


Figure 28: Loss modulus of PP/rCF (10 wt.%) 3mm_US composites before (red) and after (blue) the accelerated aging.

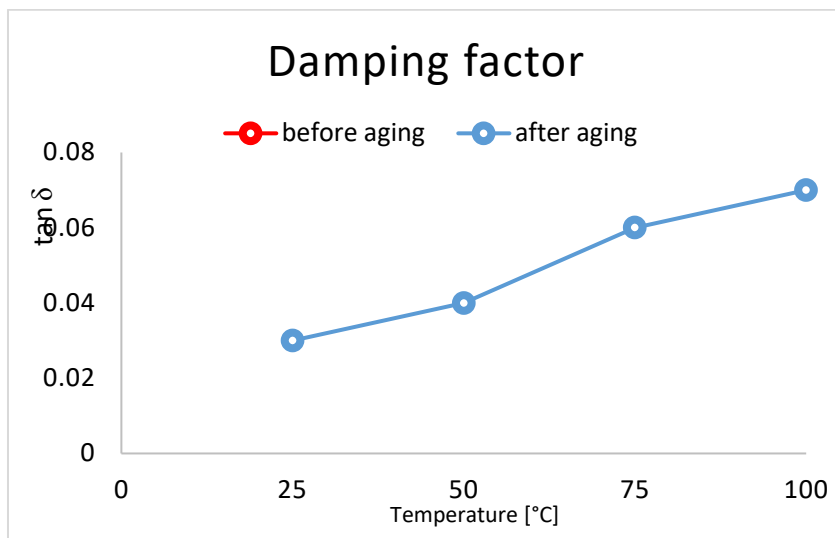


Figure 29: Tan δ of PP/rCF (10 wt.%) 3mm_US composites before (red) and after (blue) the accelerated aging.

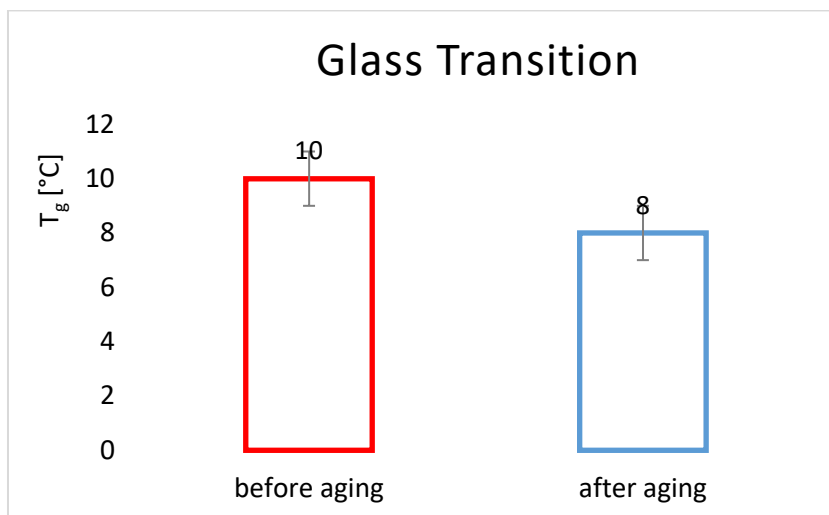


Figure 30: Glass transition temperature of PP/rCF (10 wt.%) 3mm_US composites before (red) and after (blue) the accelerated aging.

4. RESULTS OF CHARACTERIZATION OF COMPOSITES OBTAINED FROM RECYCLING OF PROTOTYPES

4.1. Materials

As above mentioned, prototypes manufactured within the project were in the form of glove compartments (Figure 9) and composed by the following composite materials:

- *vPP/rCF* virgin polypropylene reinforced with 10 wt.% of recycled carbon fibers;
- *vPA6/rCF* virgin polyamide 6 reinforced with 10 wt.% of recycled carbon fibers;

- *rPP/rCF* recycled polypropylene reinforced with 10 wt.% of recycled carbon fibers;
- *rPA6/rCF* recycled polyamide 6 reinforced with 10 wt.% of recycled carbon fibers.

In all the aforementioned cases the recycled carbon fibers are previously submitted to a sizing treatment in order to improve the adhesion with thermoplastics.

Prototypes were submitted to accelerated aging and recycling process, as described in detail in section 1.4. Then, specimen for characterization were produced.

Alongside the prototypes obtained by implementing the recycling process developed in ENEA Brindisi Research Centre laboratories, some other prototypes were obtained by substituting the compression moulding with the injection moulding step. For more clarity, in the following we will refer to the two processes with the terms Recycling process 1 and 2, respectively:

Recycling process 1:

Accelerated aging → Shredding & Milling → Extrusion → Compression Moulding

Recycling process 2:

Accelerated aging → Shredding & Milling → Extrusion → Injection Moulding

Based on this difference, the recycled materials are labelled with the suffix CM or IM, if they are produced with the recycling process 1 or 2, respectively. More in detail, the Table 8 summarizes the samples obtained via recycling process:

	Matrix	Reinforcement	Mass fraction of reinforcement	Recycling process
vPP/rCF_CM	Virgin PP	Recycled carbon fiber	10 wt.%	1
vPA6/rCF_CM	Virgin PA6	Recycled carbon fiber	10 wt.%	1
rPP/rCF_CM	Recycled PP	Recycled carbon fiber	10 wt.%	1
rPA6/rCF_CM	Recycled PA6	Recycled carbon fiber	10 wt.%	1
vPP/rCF_IM	Virgin PP	Recycled carbon fiber	10 wt.%	2
vPA6/rCF_IM	Virgin PA6	Recycled carbon fiber	10 wt.%	2
rPP/rCF_IM	Recycled PP	Recycled carbon fiber	10 wt.%	2
rPA6/rCF_IM	Recycled PA6	Recycled carbon fiber	10 wt.%	2

Table 8. Composite materials manufactured via recycling process.

Because of difficulties in extruding the granulated materials obtained by milling the prototypes made with vPP/rCF, no specimens of vPP/rCF_IM were manufactured and specimen of vPP/rCF_CM were produced by compression moulding of granulated materials (therefore excluding the extrusion step). Moreover, because of a failure in the mold temperature control system occurred in the last period of experimental trials and the inability to solve it within the end of the project, no specimens in vPA6/rCF_IM were manufactured.

For all the manufactured specimens, the tensile and impact tests, as well as the DMA, were performed. When available, the results were compared to those related to initial and aged composites, in order to study the influence of both accelerated aging and recycling process. Results are summarized in the next paragraphs.

4.2. Fiber length measurement

The measurement of the average fiber length in both initial prototypes and composites materials obtained with the recycling process 1 was performed, according to the methodology described in section 2.2.

It is known that mechanical properties of composite materials with fiber reinforcement strongly depends from the fibers length. Based on this evidence, the main aim of this measurement was to estimate the influence of recycling process on the variation of the average carbon fibers length and, therefore, on mechanical properties of recycled composites.

Samples taken from Initial composites were in the form of small pieces, cut from prototypes, and having average size of 2 cm², meanwhile the samples taken from recycled composites were in the form of extruded materials.

After the calcination, the samples taken from both initial and recycled rPA6/rCF prototypes appeared to be contaminated by traces of thermoplastic material not completely removed; as a consequence, the residuals of calcination where composed by areas of aggregated fibers and areas where fibers were more clearly separated. Only the latter were used for image processing and measurements of fibers length.

In Figure 31 an example of capture images with the light microscope and of the identification of fibers is reported.

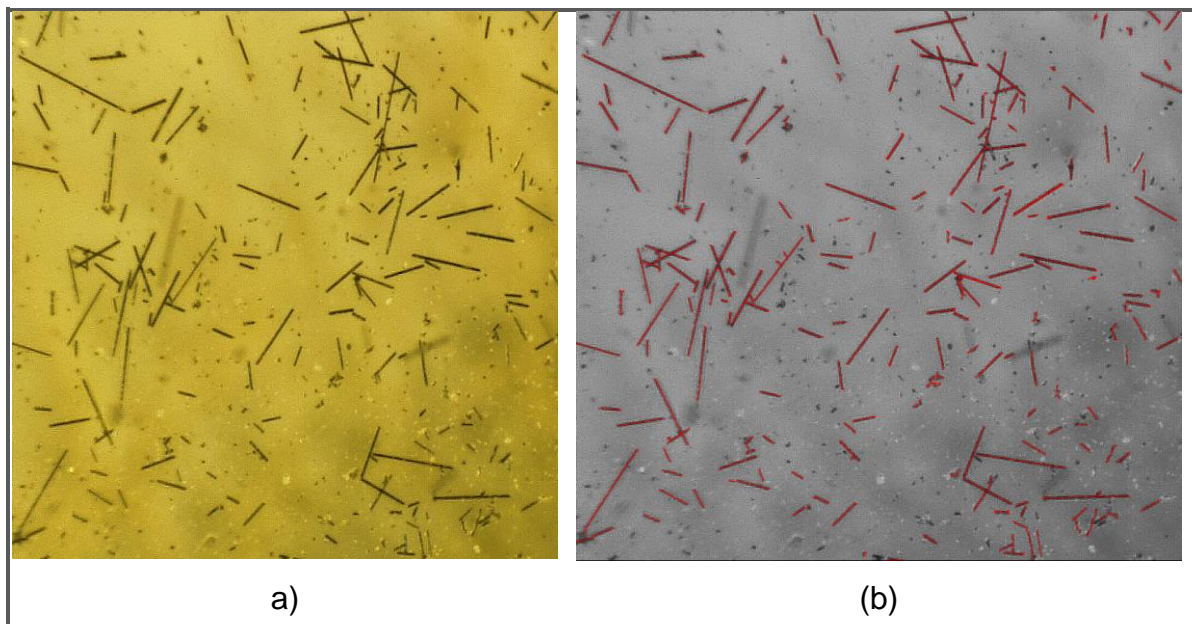


Figure 31: Image acquired from light microscope with resolution 1.82 μm (a) and fiber identification (b).

Table 9 summarizes the results of the fiber length measurements of all the composite materials investigated. From both the Figure 31 and Table 9 it appears evident that the range comprising the lengths of all the fibers is very wide, with large differences between the longer and shorter fibers, resulting in large values of standard deviations.

Considering this evidence and the similar values of the average length of the fibers before and after the recycling, it can be stated that the recycling process does not significantly affect the geometry of fibers, preserving their initial length.

		Number of fibers	Average length [μm]	Std [μm]
vPP/rCF	<i>Initial</i>	303	85.91	70.41
	<i>Recycling process 1</i>	472	98.66	84.90
vPA6/rCF	<i>Initial</i>	399	83.95	52.98
	<i>Recycling process 1</i>	343	99.62	59.93
rPP/rCF	<i>Initial</i>	533	111.25	89.74
	<i>Recycling process 1</i>	357	122.87	69.36
rPA6/rCF	<i>Initial</i>	71	91.71	60.11
	<i>Recycling process 1</i>	233	89.45	65.88

Table 9: Measurement of average length of fibers before and after the recycling process.

4.3. Tensile test

In Table 10, the results of the tensile test are reported (in the brackets, the standard deviations).

		Young's module [GPa]	Tensile strength [MPa]	Yield stress [MPa]	Strain at break (%)
vPP/rCF	<i>Initial</i>	5.5 (0.1)	34.7 (1.2)	31.1 (1.8)	1.5 (0.0)
	<i>Recycling process 1</i>	3.1 (0.5)	12.6 (1.3)	13.4 (0.0)	0.5 (0.1)
	<i>Recycling process 2</i>	-	-	-	-
vPA6/rCF	<i>Initial</i>	7.1 (0.7)	83.4 (1.7)	48.2 (8.9)	2.6 (0.2)
	<i>Recycling process 1</i>	6.8 (0.3)	92.1 (1.5)	62.2 (2.9)	6.8 (0.8)
	<i>Recycling process 2</i>	-	-	-	-
rPP/rCF	<i>Initial</i>	4.3 (0.1)	26.7 (0.7)	27.9 (0.2)	6.7 (1.0)
	<i>Recycling process 1</i>	3.4 (0.5)	19.7 (1.6)	14.9 (2.3)	2.1 (0.6)
	<i>Recycling process 2</i>	4.7 (0.0)	20.1 (0.3)	14.5 (0.5)	7.0 (0.4)
rPA6/rCF	<i>Initial</i>	4.9 (0.2)	116.1 (0.9)	116.1 (0.9)	5.3 (0.3)
	<i>Recycling process 1</i>	5.9 (1.3)	76.7 (8.8)	51.3 (5.6)	5.4 (1.1)
	<i>Recycling process 2</i>	5.2 (0.1)	64.4 (0.8)	38.7 (0.4)	5.7 (0.3)

Table 10: Results of tensile test. In the brackets, the standard deviations.

Figures 33 to 36 depict the comparison between the results of tensile test performed on initial prototypes and recycled composites manufactured with both the recycling processes 1 and 2. The comparison is done based on the following scheme:

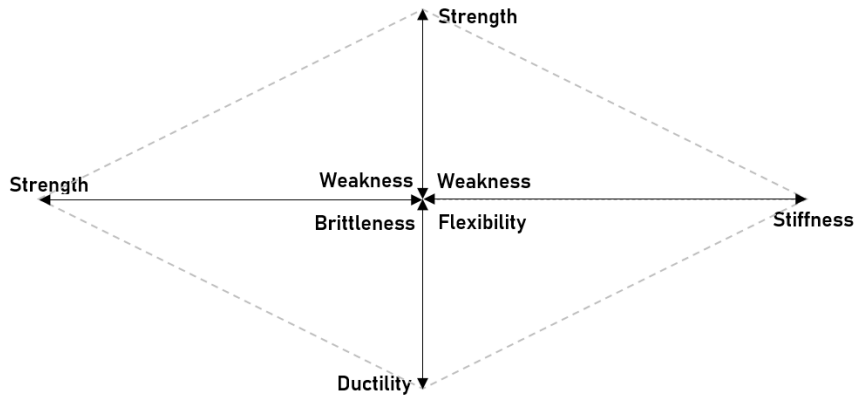


Figure 32: Scheme of mechanical properties.

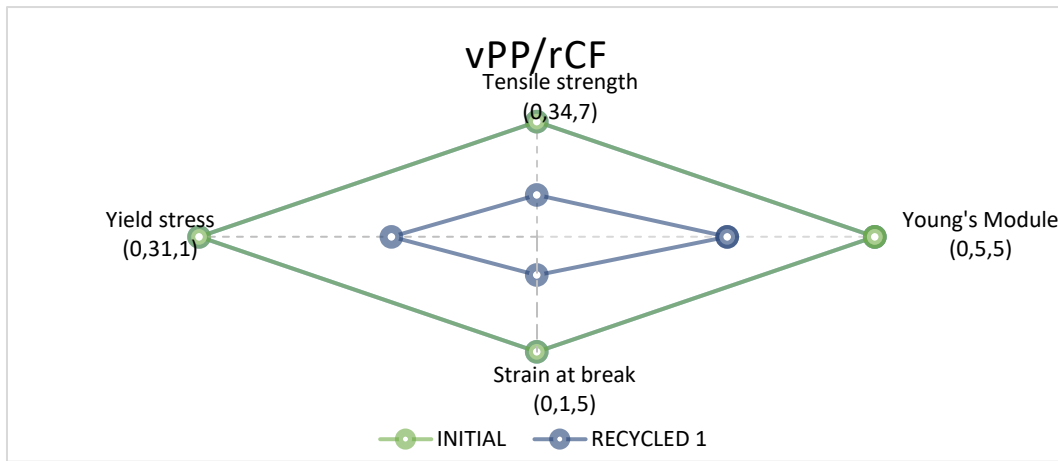


Figure 33: Tensile test on initial and recycled vPP/rCF composites.

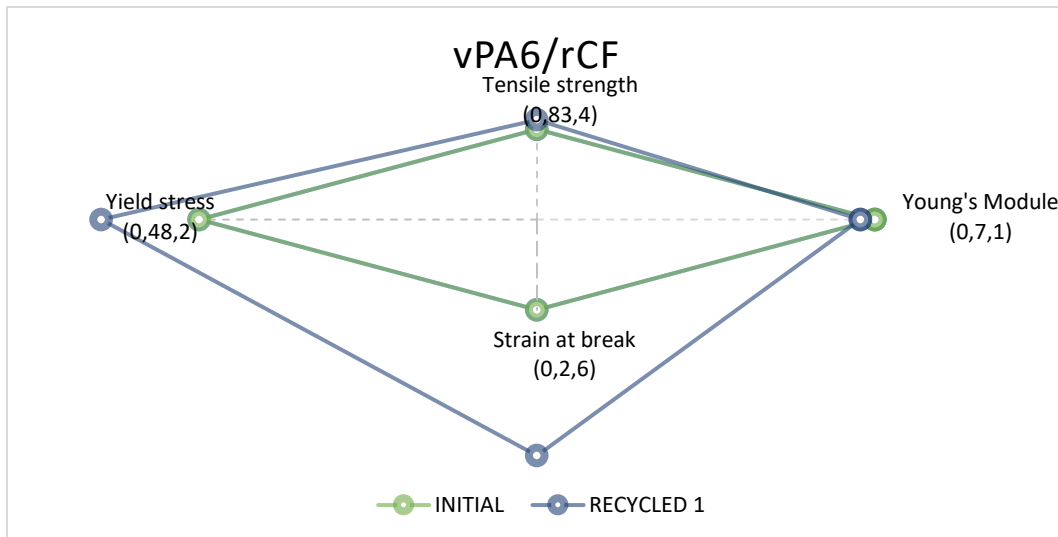


Figure 34: Tensile test on initial and recycled vPA6/rCF composites.

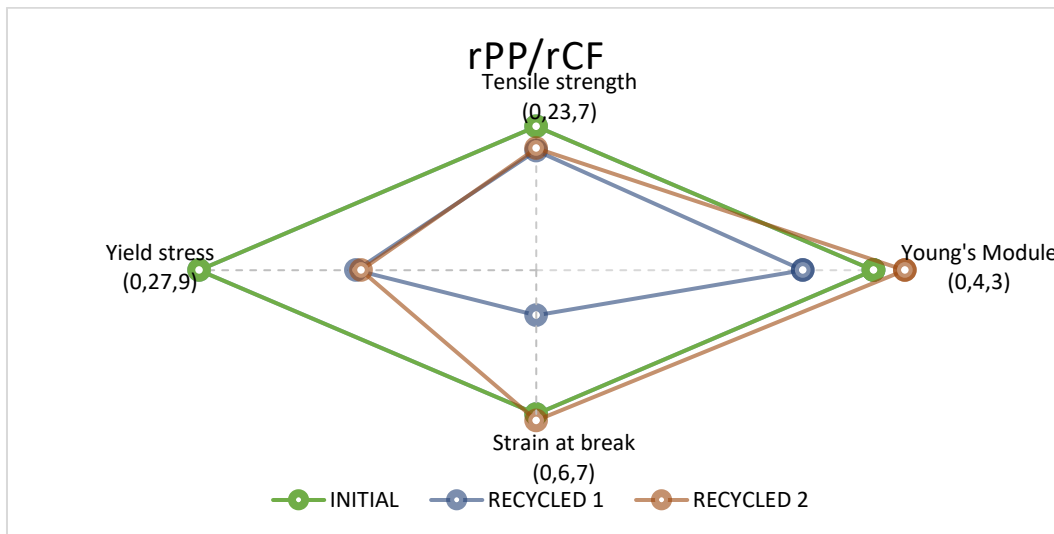


Figure 35: Tensile test on initial and recycled rPP/rCF composites.

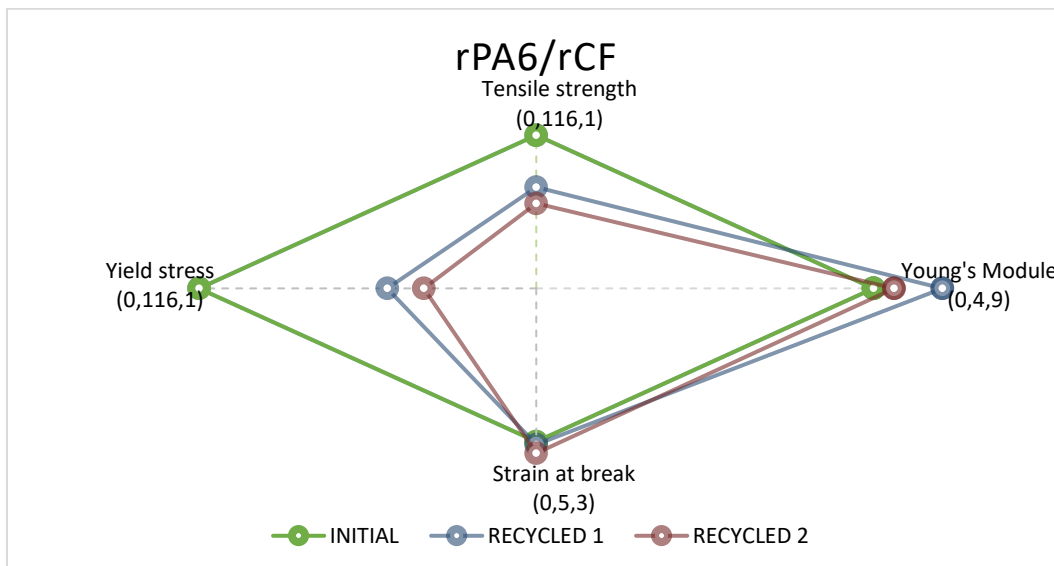


Figure 36: Tensile test on initial and recycled rPA6/rCF composites.

Figures 33 to 36 show that for the investigated materials both the tensile strength and the yield stress of recycled composites are lower than those referred to initial composites, indicating that the material has weakened due to the recycling processes 1 or 2. The reduction is more evident for composites made in vPP/rCF, followed by those in rPP/rCF and rPA6/rCF. An exception is the composite in vPA6/rCF, for which an increase in the tensile strength and especially in the yield stress values are noticed. In addition, the tensile strength and the yield stress are the same (for rPP/rCF) or a little higher (for rPA6/rCF) when composites are manufactured with the compression moulding technology rather than the injection moulding technology.

With regard to the Young's module, less clear results were obtained. When the recycling process 1 is considered, it can be stated that the Young's module decrease for vPP/rCF and rPP/rCF, it is about unchanged for vPA6/rCF and is slightly increased for rPA6/rCF. Considering this, one can state that the polypropylene seems more sensitive to the recycling process. Conversely, the Young's module appears unchanged when the recycling process 2 is considered.

Lastly, for both the virgin and recycled polypropylene, the elongation at break is strongly reduced after the recycling process 1, confirming again that this polymer is more sensitive to the recycling. Anyway, when the injection moulding technology is adopted, the elongation at break of composites made with recycled polypropylene looks unchanged. With regard to polyamide6, the elongation at break again looks unchanged when both recycling process 1 and 2 is applied on rPA6/rCF. On the contrary, beyond expected, it is strongly increased for vPA6/rCF.

4.4. Impact test

The Charpy impact test has been performed on notched specimens of initial composites as well as on un-notched specimens of aged and recycled composites. Commonly, the un-notched impact resistance of a material may be one or two orders of magnitude higher than the notched impact resistance for the same material. Based on this general statement, no direct comparison is possible among results reported in Table 11; nevertheless, it can be likely inferred that in all the cases the Charpy impact values for the initial composites are greater than those related to the aged and recycled ones.

		Charpy Impact value* [kJ·m⁻²]
vPP/rCF	<i>Initial</i>	-
	<i>Aged</i>	3.7 (1.5)
	<i>Recycling process 1</i>	7.0 (1.5)
	<i>Recycling process 2</i>	-
vPA6/rCF	<i>Initial</i>	3.3 (0.5)
	<i>Aged</i>	18.2 (10.4)
	<i>Recycling process 1</i>	51.5 (10.5)
	<i>Recycling process 2</i>	-
rPP/rCF	<i>Initial</i>	3.7 (0.2)
	<i>Aged</i>	36.5 (0.3)

	<i>Recycling process 1</i>	18.8 (2.8)
	<i>Recycling process 2</i>	38.0 (3.0)
rPA6/rCF	<i>Initial</i>	3.2 (0.4)
	<i>Aged</i>	59.2 (1.1)
	<i>Recycling process 1</i>	23.2 (5.9)
	<i>Recycling process 2</i>	49.0 (5.8)

** Notched specimens were used for initial composites; un-notched specimens were used for recycled composites.*

Table 11: Results of Charpy impact test of composites. In the brackets, the standard deviations.

In particular, a comprehensive study of impact resistance has been performed for rPP/rCF and rPA6/rCF, for which values referred to initial, aged, recycling process 1 and recycling process 2 are available, and relative results are depicted in Figure 37. Considering that the notched impact resistance is from one to two orders of magnitude lower than the un-notched impact resistance, from Figure 37 is reasonable to infer that impact resistance of aged composites is lower than that of initial composites. Recycling process contributes to further reduce the impact resistance, in particular for recycling process 1, which seems more affecting in this sense when compared to recycling process 2. Indeed, in particular for rPP/rCF composites, the impact resistance of materials submitted to recycling process 2 is very similar to that of aged composites.

Lastly, as for the tensile test, also for the impact test the composites made with polypropylene seems more sensitive to aging and recycling processes. Indeed, although the impact resistance of initial composites in rPP/rCF is a little higher than that of rPA6/rCF, the opposite happens after the aging and the recycling process, being the values for composites in rPP/rCF lower than those referred to PA6/rCF.

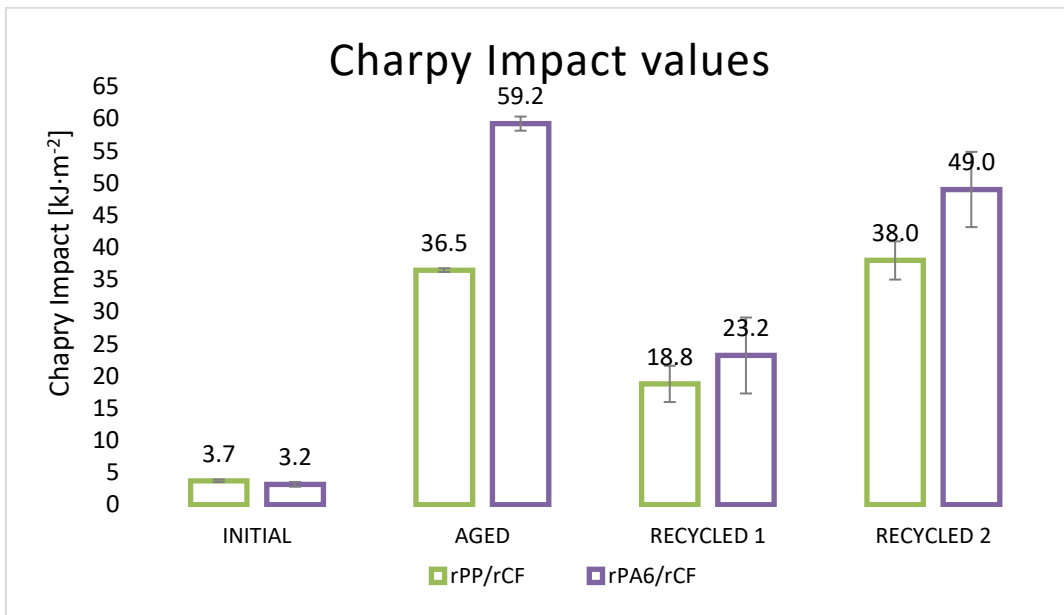


Figure 37: Result of Charpy impact test on composites based on recycled polymers.

4.5. Dynamic Mechanical Analysis

In Table 12 the storage, the loss modulus and the damping factor values on initial, aged and recycled composites were collected. With regard to recycling process, the DMA was carried out only on samples manufactured with the recycling process 1, including the compression moulding step.

Temperature [°C]	Storage Modulus [MPa]						Loss Modulus [MPa]						Damping Factor					
	30	60	90	120	30	60	90	120	30	60	90	120	30	60	90	120		
VPA6/rCF	Initial	2769 (365)	1960 (237)	1218 (115)	861 (74)	97 (12)	142 (19)	67 (6)	35 (3)	0.04 (0.01)	0.07 (0.01)	0.05 (0.01)	0.04 (0.01)	0.07 (0.01)	0.05 (0.01)	0.04 (0.00)		
	Aged	2729 (254)	1931 (313)	1298 (149)	1068 (88)	110 (8)	141 (26)	65 (13)	33 (3)	0.04 (0.00)	0.07 (0.00)	0.05 (0.00)	0.04 (0.00)	0.07 (0.00)	0.05 (0.00)	0.03 (0.00)		
	Recycling process 1	3606 (89)	2829 (89)	1429 (64)	967 (48)	56 (12)	217 (3)	121 (4)	51 (2)	0.02 (0.01)	0.08 (0.01)	0.08 (0.01)	0.02 (0.01)	0.08 (0.01)	0.08 (0.01)	0.05 (0.00)		
rPA6/rCF	Initial	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-		
	Aged	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-		
	Recycling process 1	3768 (69)	2735 (17)	1447 (16)	978 (15)	90 (2)	247 (3)	126 (2)	57 (1)	0.02 (0.00)	0.09 (0.00)	0.09 (0.00)	0.02 (0.00)	0.09 (0.00)	0.09 (0.00)	0.06 (0.00)		
rPP/rCF	Initial	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-		
	Aged	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-		
	Recycling process 1	2565 (103)	1875 (84)	1300 (118)	909 (92)	103 (7)	95 (6)	100 (7)	85 (8)	0.04 (0.00)	0.05 (0.00)	0.08 (0.00)	0.04 (0.00)	0.05 (0.00)	0.08 (0.00)	0.09 (0.00)		
rPP/rCF	Initial	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-		
	Aged	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-		
	Recycling process 1	2981 (143)	2325 (110)	1685 (85)	1108 (69)	131 (14)	125 (811)	141 (12)	127 (11)	0.04 (0.01)	0.05 (0.01)	0.08 (0.01)	0.04 (0.01)	0.05 (0.01)	0.08 (0.01)	0.1 (0.01)		

Table 12: Storage, loss modulus and damping factor values on initial, aged and recycled PA6/rCF and PP/rCF composites. In the brackets, the standard deviations.

The comparison of the storage, loss and damping factor of initial, aged and recycled vPA6/rCF composites was reported in Figures 38 to 41. The recycled 1 vPA6/rCF composites were found more stiff than the others samples, and the loss modulus was found higher due to reduced mobility. Conversely, a higher damping factor was found and it was generally associated to a poor fibre-matrix adhesion.

The DMA study indicated that the increased modulus, together with the increased in $\tan \delta$ values on recycled 1 composites, was attributed to wickered interaction between the polymer and carbon fibers not able to restrict the segmental mobility of the polymer chains and to promote the thermal stability to the composites.

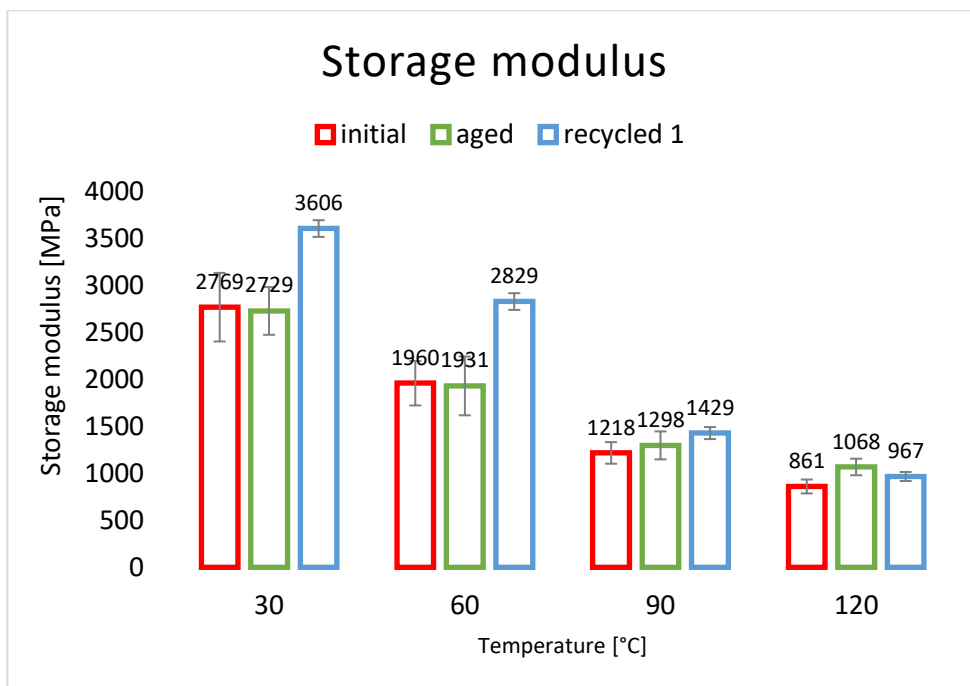


Figure 38: Storage modulus of initial, aged and recycled vPA6/rCF composites.

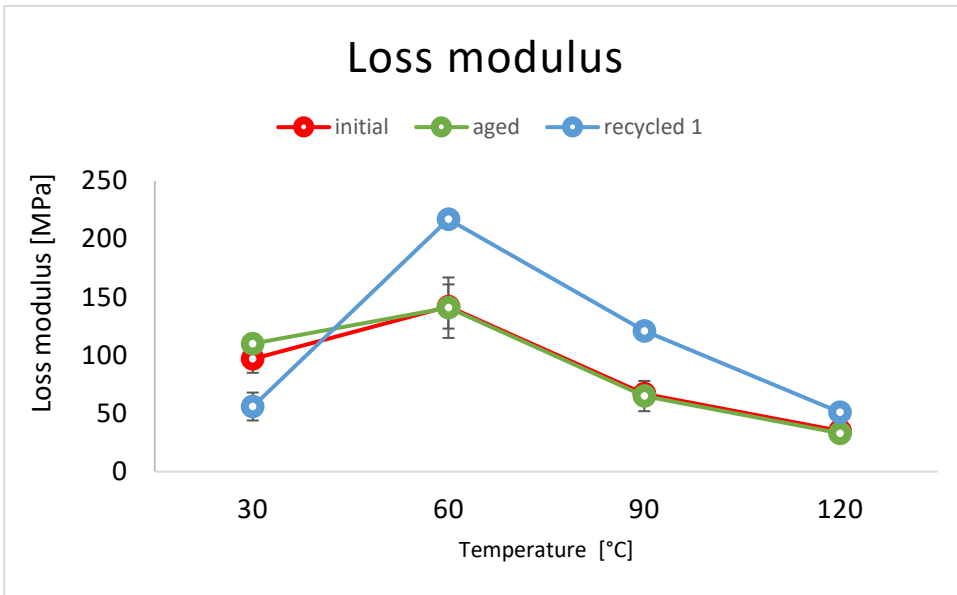


Figure 39: Loss modulus of initial, aged and recycled vPA6/rCF composites.

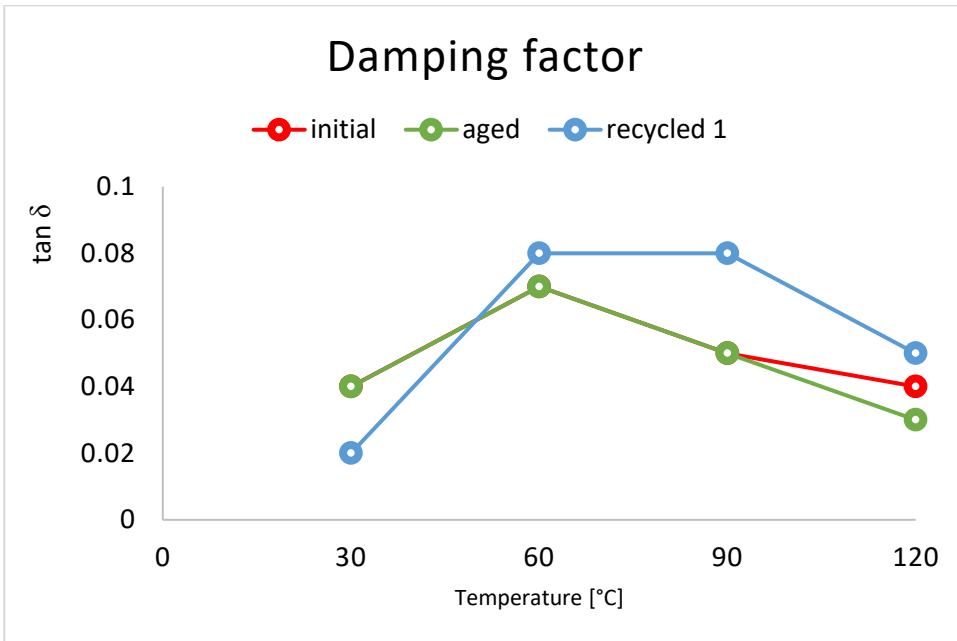


Figure 40: Tan δ of initial, aged and recycled vPA6/rCF composites.

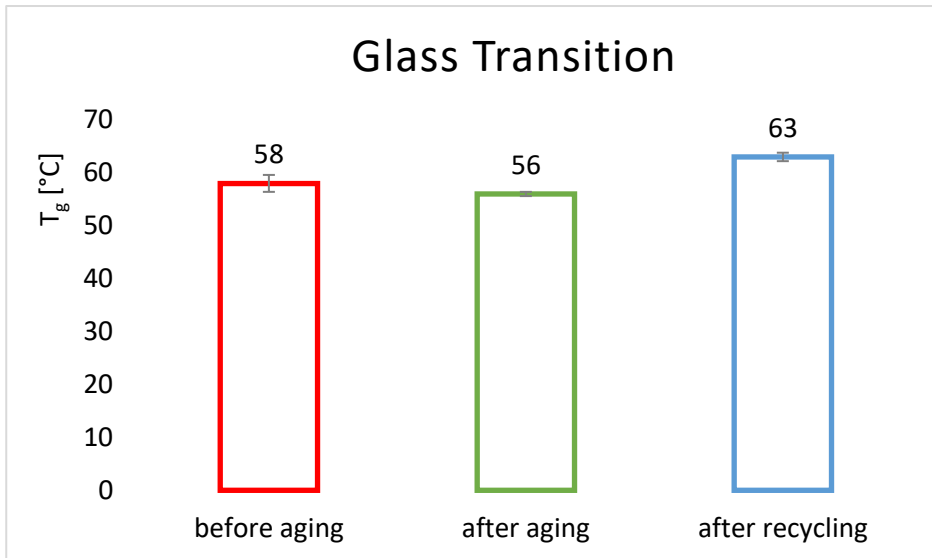


Figure 41: Glass transition temperature of initial, aged and recycled vPA6/rCF composites.

5. CONCLUSIONS

The first part of this document summarizes the activities focused on the development of a mechanical recycling process of composite materials made with virgin and recycled thermoplastics (PP or PA6) and recycled carbon fibers. In particular, the recycled carbon fibers come from pyrolysis of dismissed composite based components. The second part is devoted to the characterization and analysis of both intermediate and final products manufactured by implementing the developed recycling process, with the aim to verify its effectiveness in view of possible market exploitation.

The recycling process consists in a preliminary accelerated weathering of prototypes, to simulate natural weathering they experiment during lifetime, followed by shredding & milling, extrusion and moulding steps.

In ENEA laboratories, the recycling process was first successfully applied on preliminary samples, in order to roughly determine the process parameters. Then, it was applied on prototypes manufactured within the REVALUE project: more in detail, they were glove compartments intended for Automotive sector and made with virgin or recycled Polypropylene and Polyamide 6, reinforced with 10 wt.% of recycled carbon fiber. It was noticed that vPA6/rCF could be processed without impediments, to manufacture composite plates. Conversely, despite of wide range of tested process parameters, the extrusion of vPP/rCF did not work and materials could not be extruded. The most plausible hypothesis is that the accelerated weathering caused a degradation of polypropylene that compromised the processability.

After these preliminary tests, the recycling process has been successfully replicated and when necessary optimized on prototypes developed within the project. Contextually, mass and energy balances of the overall process were carried out. They pointed out that negligible losses of materials are observed in the shredding/milling and extrusion steps, but conversely a greater loss of materials occurs in the compression moulding, due to deflashing of moulded. Nevertheless, the overall yields of recycling process for vPA6/rCF (10 wt.%)_S is quite high: 88.4 %. With concerning to the energy balance, it can be stated that the more demanding step is the compression moulding, with values a little higher that those commonly referred to compression moulding of CFRP. However, the overall energy consumption is estimated to be in line with the commonly known industrial data and the specific energy consumption (SEC) is in the order of 16-17 MJ·kg⁻¹.

Measurements of fiber length before and after the recycling process was performed, with the aim to understand at what extent the recycling process affects the average lengths of

the reinforcement. Moreover, aged and recycled composites were characterized with tensile, impact tests and DMA analysis.

From fiber length measurement it can be stated that the recycling process does not significantly affect the average length of fibers, preserving their initial values. Nevertheless, it is worth notice that the initial composites were already manufactured with recycled carbon fibers having a very short average length that was not further reduced during the shredding and milling of materials. This contributed to reduce to a lower extent the mechanical properties of the composites after the overall recycling process.

From tensile test results, one can state that the polypropylene seems more sensitive to the recycling process, for which lower values of both tensile strength and yield stress were observed. Similarly, the Young's module decreases as effect of recycling process 1 (employing compression moulding), in particular for composites made with polypropylene; on the contrary, the Young's module appears unchanged when the recycling process 2 (employing injection moulding) is applied.

Aging and recycling processes also contribute to reduce the impact resistance of composites, in particular when compression moulding technology is applied. Even from impact test results it can be stated that composites made with polypropylene seems more sensitive to aging and recycling processes, their impact resistance values being reduced to a greater extent than those referred to polyamide 6-based composites.

Dynamic mechanical analysis shows that the aging process did not significantly affect neither the stiffness nor the viscous nature of both sized and unsized composites made with polyamide 6, in the entire temperature range explored. Conversely, the aging increasing the loss modulus of unsized PP/rCF compounds. Moreover, for both the thermoplastic matrices no effects of aging on the damping factor were found.

On the other hand, DMA results depict that the recycling process seems to be more affecting. Indeed the recycled vPA6/rCF composites were found more stiff than both initial and aged ones, while the loss modulus was found higher due to reduced mobility. In addition, an increase of damping factor was found after the recycling. The increased modulus, together with the increased in $\tan \delta$ values on recycled composites, was attributed to wicker interaction between the polymer and carbon fibers not able to restrict the segmental mobility of the polymer chains and to promote the thermal stability to the composites.

In conclusion even if, as expected, the recycling process affects the overall performance of the composites, some positive conclusions can be drawn from our investigation. In fact the mechanical properties retained in particular by recycled PA6/rCF based composites allow

the potential use of recycled materials in applications as automotive semi-structural or non-structural parts of equipment housing (like car door panel, car headlamp reflectors, etc).

6. REFERENCES

1. Web site:

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2. Howart J., Mareddy S.S.R., Mativenga P.T., Energy intensity and environmental analysis of mechanical recycling of carbon fibre composite, Journal of Cleaner Production, 81 (2014) 46–50. [Http://dx.doi.org/10.1016/j.jclepro.2014.06.023](http://dx.doi.org/10.1016/j.jclepro.2014.06.023)

ATTACHMENTS

Attachment 1 – Data sheets of polymers

Data sheet of polypropylene SABIC PP 595A



SABIC® PP 595A

POLYPROPYLENE HOMOPOLYMER FOR USE IN AUTOMOTIVE COMPOUNDING

DESCRIPTION

SABIC® PP 595A has been specially developed for use in automotive compounding. The material has high flow properties and a high stiffness, enabling high production rates. It is formulated with a dedicated automotive additive package.

TYPICAL PROPERTY VALUES

Revision 20170706

PROPERTIES	TYPICAL VALUES	UNITS	TEST METHODS
POLYMER PROPERTIES			
Melt Flow Rate			
at 230 °C and 2.16 kg	47	dg/min	ISO 1133
Density at 23°C	905	kg/m ³	ASTM D1505
MECHANICAL PROPERTIES			
Tensile Strength at Yield ⁽¹⁾	35	MPa	ASTM D638
Tensile Elongation at Yield	11	%	ISO 527-1/-2
Flexural Modulus (1% Secant)	1800	MPa	ASTM D790 A
Notched Izod Impact Strength at 23°C	20	J/m	ASTM D256
Rockwell Hardness, R-Scale	104	-	ASTM D785
THERMAL PROPERTIES			
Vicat Softening Point	152	°C	ASTM D1525
Heat Deflection Temperature at 455kPa	108	°C	ASTM D648

(1) Based on injection molded specimens

STORAGE AND HANDLING

Polypropylene resin should be stored in a manner to prevent a direct exposure to sunlight and/or heat. The storage area should also be dry and preferably do not exceed 50°C. SABIC would not give warranty to bad storage conditions which may lead to quality deterioration such as color change, bad smell and inadequate product performance. It is advisable to process PP resin within 6 months after delivery.

DISCLAIMER

The information contained herein may include typical properties of our products or their typical performances when used in certain typical applications. Actual properties of our products, in particular when used in conjunction with any third party material(s) or for any non-typical applications, may differ from typical properties. It is the customer's responsibility to inspect and test our product(s) in order to satisfy itself as to the suitability of the product(s) for its and its customers particular purposes. The customer is responsible for the appropriate, safe and legal use, processing and handling of all product(s) purchased from us. Nothing herein is intended to be nor shall it constitute a warranty whatsoever, in particular, warranty of merchantability or fitness for a particular purpose.

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CHEMISTRY THAT MATTERS™

Data sheet of polyamide 6 RADILON S27 100NT



PERFORMANCE PLASTICS



TECHNICAL DATA SHEET

RADILON S 27 100 NT

Material code Colour code

PROPERTY	STANDARD	UNIT	VALUE	
			DAM*	Cond**
Physical Properties				
Density	ISO 1183	Kg/m ³	1140	
Moisture absorption 23°C - 50%RH	2mm thk ISO 62	%	2.7	
Water absorption, immersion at 23°C	2mm thk ISO 62	%	10.5	
Viscosity Index (Sulfuric Acid)	ISO 307	ml/g	145	
Mechanical Properties				
Tensile Modulus	1mm/min ISO 527-2/1A	MPa	2900	1100
Stress at Yield	50mm/min ISO 527-2/1A	MPa	70	40
Yield Strain	50mm/min ISO 527-2/1A	%	4	30
Nominal Strain at Break	50mm/min ISO 527-2/1A	%	70	>50
Flexural Modulus	2mm/min ISO 178	MPa	2300	750
Flexural Strength	2mm/min ISO 178	MPa	95	30
Charpy Notched Impact Strength	+23°C ISO 179/1 eA	KJ/m ²	4	
Charpy Notched Impact Strength	-30°C ISO 179/1 eA	KJ/m ²	3	
Thermal Properties				
Melting Temperature	10°C/min ISO 11357-1-3	°C	220	
Heat Deflection Temperature	1.8 MPa ISO 75/2Af	°C	55	
Vicat Softening Temperature	50°C/h ISO 308/B50 50N	°C	195	
Flammability Properties				
Flammability	0.8mm UL 94	class	HB	
Glow Wire Flammability Index	2mm IEC 60695-2-12	°C	700	
Automotive interior flammability	3mm thk ISO 3795	mm/min	0	
Electrical Properties				
Volume resistivity	500V IEC 60093	ohm · m	1 E+13	1 E+11
Surface resistivity	500V IEC 60093	ohm	1 E+12	1 E+10
Comperative Trecking Index	SoLA IEC 60112	-	600	

*DAM = Dry As Moulded state **Cond = Conditioned state similar to ISO 1110 ***Melt Temp [°C] / Mold Temp [°C] / Cavity press [MPa]

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The information provided in this documentation corresponds to knowledge of Radici Group Performance Plastics on the subject at the date of its publication. This information may be subject to revision as new knowledge and experience become available. The data provided reflects the average values of the properties measured over an adequate number of different production cycles and relates only to the designated material; this data may not be valid for each material used in combination with any other materials or additive or in any process, unless expressly indicated otherwise. The data provided should not be used to establish specification limits nor used alone as the basis of design; it is not intended to substitute for any testing you may need to conduct to determine for yourself the suitability of a specific material for your particular purpose. Since Radici Group Performance Plastics cannot anticipate all variations in actual end-use conditions Radici Group Performance Plastics makes no warranty and assumes no liability in connection with any use of this information. Nothing in this publication is to be considered as a license to operate under or a recommendation to infringe any patent rights.

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