Static mechanical properties of virgin and recycled short glass fiber-reinforced polypropylene produced by pellet additive manufacturing

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Abstract
The static behavior of specimens made of virgin and recycled short glass fiber-reinforced polypropylene filled with mineral filler has been studied. Specimens were produced by means of pellet additive manufacturing (PAM) adopting different infill patterns to investigate the influence of material anisotropy. For comparison purposes, the mechanical properties of the same virgin and recycled materials were investigated also with injection-molded specimens. The fracture behavior was investigated by analyzing the damage at a macroscopic as well as microscopic level. As the main outcome, it was noted that both the manufacturing and recycling processes play a crucial role in the damage mechanics of the specimens, thus explaining the different fracture behaviors noted at the macroscale.

Keywords
additive materials, fracture surface, polymer alloy, tensile strength

1 | INTRODUCTION

Polymers and short fiber-reinforced polymer composites (SFRPCs) are groups of materials widely adopted in the additive manufacturing (AM) technology, thanks to their excellent physical and chemical properties combined with adequate mechanical properties for structural applications. However, the downside of this large consumption is the large amount of waste, leading to deleterious environmental, health, and economic impacts. Nowadays, recycling is a pivotal aspect to reduce product costs as well as the environmental impact of plastics. Along with recycling, design approaches based on topology optimization can be synergically used to reduce the waste of plastics taking advantage of AM technologies. For these reasons, recycled polymer materials for 3D printing became recently available, such as recycled linear low-density polyethylene (rLLDPE), acrylonitrile butadiene styrene (rABS), polycarbonate (rPC), polypropylene (rPP), polyethylene terephthalate (rPET), or polylactic acid (rPLA).

A remarkable conclusion of the aforementioned papers is the positive technical feasibility to use recycled polymers for 3D printing process, but not all polymers have reached the same maturity in terms of mechanical performance, as compared with the relevant virgin material or with the injection molding (IM) process. Up to now, the best results were achieved by using rABS and rPLA. Czyżewski et al. compared the mechanical properties of injection-molded and 3D printed...
specimens made of rABS, obtained by fused filament fabrication (FFF) also called fused deposition modeling (FDM), and they found a decrease in the elastic modulus (12.9%), in the ultimate tensile strength (19.5%), and in the ultimate tensile strain (33.1%) of 3D printed specimens with respect to those obtained by IM, due to the occurrence of discontinuity areas in the volume of the FDM samples that depends on the process parameters (especially temperature), on the recycling process, and on the continuous filament production. Mohammed et al. investigated the changes in physical and mechanical properties of ABS manufactured using FDM 3D printing, across two recycling phases. They found that rABS showed a reduction in both tensile (25.6%) and compressive (26.3%) strengths compared to the virgin material. However, compared to one-time rABS, two time rABS showed increased strength, implying that the improved recycled state of the polymer may be advantageous for the resulting mechanical properties and consequently potential manufacturing applications. The reduction in tensile and compressive strength of the horizontally oriented specimens was limited to 16.8% and 16.9%, respectively, compared to the virgin material. Similar results were found by Vidakis et al. who investigated the influence of repeating recycle processes on the mechanical properties of rABS FDM specimens. Standard tensile, compression, flexural, impact, and microhardness tests were carried out, and it was found that the mechanical properties of rABS polymer are generally improved with the repetition of recycling cycles, an optimum overall mechanical behavior having been found between the third and fifth repetitions. A degradation of mechanical properties was found after the fifth recycling cycle.

Concerning PLA, Anderson compared the mechanical properties of 3D printed virgin and recycled specimens, carrying out tensile, shear, and hardness tests. It was found that with the recycled filament, the elastic modulus was statistically unchanged and the shear strength increased by 6.8%, while the tensile strength and hardness decreased by 10.9% and 2.4%, respectively. Although the average mechanical properties before and after recycling were similar, there was more variability in the results obtained with the recycled filament. Similar results were found by Zhao et al. who analyzed the repeated FDM 3D printing process on PLA specimens and found that the 3D printing can be repeated only for two cycles, due to the significant deterioration detected in the viscosity. They noticed that the elastic modulus and the yield strength were unchanged, while the ultimate tensile strength and elongation at break decreased by 2.4% and 9.8%, respectively.

Promising materials are also PP and PET, provided that additional efforts are made to improve their printability due to the weak adhesion to the base plate, the high warping/shrinking tendency, and the weak interfacial welding between printed layers. A possible way is blending recycled PP and polystyrene (PS), as highlighted by Zander et al. In fact, the authors compared the tensile strength of rPP specimens obtained by IM and by FDM 3D printing process. In the latter case, the specimens were obtained using both rPP and a rPP/rPS 50–50 blend, and it was found that the tensile strength decreased by 41.7% and 33.3% when using rPP and a rPP/rPS 50–50 blend, respectively.

Other strategies to improve the recycled FDM 3D printed materials consist in the incorporation of carbon, glass, or natural fibers into the polymeric filament. Tian et al. recycled continuous carbon fiber and PLA matrix in the form of PLA impregnated carbon fiber filament from 3D printed composite components and reused it as the raw material for further 3D printing process. Tensile strength of recycled carbon fiber filaments was evaluated and resulted higher than the originally printed composites, due to the improved interfacial properties. Remanufactured specimens also exhibited a 25% higher bending strength than the original ones. Rahimizadeh et al. proved the feasibility of using recycled glass fibers (GFs) from wind turbine blades with comparable mechanical properties to the virgin filament. An 8% improvement in the elastic modulus compared to pure PLA samples was observed, while there was no meaningful increase in the tensile strength.

To the best knowledge of the authors, few studies are available concerning recycled short fiber-reinforced PP composites. Stoof and Pickering analyzed the mechanical performance of composite filaments with different harakeke, hemp fiber, or recycled gypsum contents (0–50 wt%) in recycled pre-consumer PP. They found that the most successful filaments contained 30 wt% of harakeke fibers with tensile strength and elastic modulus of 39 MPa and 2.8 GPa, respectively, leading to a 74% increase in the tensile strength and 214% in the elastic modulus compared to those of plain PP filament. Veer et al. investigated the mechanical behavior of 3D printed samples by using rPP, reinforced by short GFs. The tensile strength and fracture strain were compared with literature values relevant to virgin material, and a 12.5% decrease in the tensile strength was found.

This paper analyzes the static tensile properties of virgin and recycled short GF-reinforced PP filled with a mineral filler (MF), with the following aims:

- to provide new experimental results useful for engineers involved in structural design with virgin and recycled SFRPCs and
to investigate the effect of the manufacturing process, namely, IM and pellet-based AM, on the damage mechanisms and overall strength of the considered material.

To this aim, static tensile tests were carried out on specimens manufactured by using virgin or post-consumer recycled PP manufactured by the well-established IM and more recent pellet AM (PAM) procedures. Before testing, fiber length distributions were obtained from post-manufactured specimens to quantify the level of fiber breakage during the manufacturing process. Eventually, the damage mechanisms were investigated at both the macroscopic and microscopic scales, to analyze the effects of the material and the manufacturing process.

2 MATERIALS AND TESTING METHODS

2.1 Materials and manufacturing processes

The feedstock used for producing the specimens consisted of pellets of short GF-reinforced polypropylene (PP) filled with MF. The contents of the fibers and filler were intentionally omitted due to confidentiality reasons with the company that provided the materials. One set of pellets was prepared by using virgin PP (vPPMF/GF), while post-consumer recycled PP was used for the second set (rPPMF/GF).

The specimens were produced by two different manufacturing technologies, namely, IM and PAM, also known as pellet-based filament deposition modeling process (p-FDM).

The virgin and recycled IM specimens were prepared according to the geometry reported in the ASTM D638-1420 (Figure 1A), by setting the following process parameters: (i) melt temperature ranging from 220°C and 235°C; (ii) mold filling time 0.40 s (maximum measured injection pressure 1100–1200 bar); (iii) holding time 10 s (measured holding pressure 400 bar); and (iv) cooling time 20 s. The mold-fill direction was parallel to the specimen’s longitudinal axis (Figure 2A).

Concerning the 3D printed specimens, all batches were produced by means of a Pollen® Series P 3D printer, which is a multi-material fully customizable 3D printer capable to print thermoplastics starting from pellets. Figure 1B shows the specimen’s geometry, whose longitudinal size was reduced compared to Figure 1A to avoid distortions due to the high tendency of shrinking and warping of the PP. All specimens were produced horizontally with respect to the platform, in the flat (F) position, as shown in Figure 2B, and by setting an infill density parameter equal to 100%. To ensure good adhesion between the platform and the PP matrix, the specimens were manufactured over a previously 3D printed base plate of the same material. The layer thickness was set equal to 0.3 mm, and an air gap of 0.19–0.20 mm was imposed between the base plate and the first layer of the specimen to make detachment of the specimens from the base plate easy. As a result, the roughness of the specimen’s surface corresponding to the first printed layer was lower than the opposite one (i.e., the last printed layer of the specimens).

The AM system was equipped with an extruder having 0.6-mm nozzle diameter, and the process parameters were progressively tuned for minimizing warping. The final set of parameters is summarized in Table 1.

Three sets for each AM material were manufactured by using different infill patterns defined by the angle between the filament direction and the specimen’s longitudinal axis, as schematically reported in Figure 2C–E. According to this definition, the infill patterns shown in Figure 2C–E are 0°, 90°, and ±45°, respectively. The
notation ±45° means that the filament orientation of subsequent layers changes from +45° to −45°. The infill strategy consisted of depositing first the so-called wall, that is, the contour of the profile, and then infilling the profile with the relevant infill pattern. The wall thickness was composed by one raster to reduce its influence on the mechanical properties of the specimens when the infill pattern is 90° or ±45°.

Eventually, the Fourier transform infrared (FTIR) spectroscopy was adopted to investigate the structure of vPPMF/GF and rPPMF/GF pellets. Spectra were obtained using a Thermo Nicolet iN10 MX spectrometer model paired with a Nicolet iZ10 attenuated total reflection (ATR) probe. IR spectra were obtained in the range of 4000–525 and were performed with 4-cm⁻¹ resolution using 16 scans.

### 2.2 Fiber length analysis

One sample per each material (virgin and recycled) and manufacturing process (IM and AM_F0°) was adopted to evaluate the fiber length (L). The polymer matrix was burnt off in a muffle furnace at a temperature of 550°C for 3 h. Afterwards, the fibers were cleaned by using hydrochloric acid and then filtered and dried to remove the residuals of the MF. Several images were digitally acquired using an optical microscope to obtain a total number of fibers per each material ranging from 1000 to 1400. The lengths were measured manually by processing the images acquired using the image analysis software ImageJ 1.53a.

Since the lengths were measured by excluding the fibers that intersected the contour of the picture acquired by the optical microscope, the fiber length distributions were adjusted with the method proposed by Fu et al., according to the following description. The length data were subdivided into constant width n-bin, ΔL, between the minimum and maximum fiber length measured. Let
us indicate with \( N_{i\text{,init}} \) the number of fibers measured from an image with length between \( L_i - \Delta L/2 \) and \( L_i + \Delta L/2 \), where \( L_i \) is the center of the \( i \)th bin. Fu et al.\(^{22} \) suggest correcting \( N_{i\text{,init}} \) by including the number of fibers that intersect the contours, \( N_{i\text{,int}} \), to obtain the true value of the frequency number \( N_{i\text{,lad}} \):

\[
N_{i\text{,lad}} = N_{i\text{,init}} + N_{i\text{,int}}
\]

The relation between \( N_{i\text{,init}} \) and \( N_{i\text{,int}} \) was proposed by Fu et al.,\(^{22} \) and it is described by Equation (2):

\[
\frac{N_{i\text{,int}}}{N_{i\text{,init}}} = \frac{\frac{4}{\pi} (A + B)L_i}{A \cdot B - \frac{4}{\pi} AL_i - \frac{4}{\pi} BL_i + \frac{4}{\pi} L_i^2}
\]

where \( A \) and \( B \) are the width and height of the picture, which were 4162 and 3122 \( \mu \)m, respectively.

The fiber length distribution is characterized by using the fiber length relative density \( f_{i\text{,lad}}(L_i) \):

\[
f_{i\text{,lad}}(L_i) = \frac{N_{i\text{,lad}}N_{\text{tot,lad}}}{\Delta L}
\]

where \( N_{\text{tot,lad}} \) is the summation of \( N_{i\text{,lad}} \).

From the adjusted distributions, the numeric mean and the weighted mean fiber length (\( L_{m\text{,lad}} \) and \( L_{w\text{,lad}} \), respectively) are obtained as follows:

\[
L_{m\text{,lad}} = \frac{\sum_{i=1}^{n} N_{i\text{,lad}}L_i}{\sum_{i=1}^{n} N_{i\text{,lad}}}
\]

\[
L_{w\text{,lad}} = \frac{\sum_{i=1}^{n} N_{i\text{,lad}}L_i^2}{\sum_{i=1}^{n} N_{i\text{,lad}}L_i}
\]

The raw mean \( L_m \) and weighted mean \( L_w \) values can be calculated by using \( N_i \) instead of \( N_{i\text{,lad}} \).

### 2.3 Tensile test setup and damage analysis

Displacement-controlled tensile tests were performed by using an electromechanical testing machine STEP Lab EA05 with a load capacity of 5 kN, controlled by a Test Center STEP Lab digital controller. For the IM specimens (Figure 1A), the adopted displacement rate was equal to 50 mm/min. Due to the different length between IM and AM specimens, to maintain approximately the same strain rate, the tensile tests on AM specimens (Figure 1B) were performed at a reduced rate of 25 mm/min. The strain was measured by using an MTS extensometer having a gauge length equal to 25 mm. The resulting strain rates measured by the extensometer in the linear range were approximately \( 4.8 \times 10^{-3} \) and \( 4.6 \times 10^{-3} \) s\(^{-1} \) for IM and AM specimens, respectively. The material elastic modulus, \( E \), the tensile strength, \( \sigma_m \), and the elongation at break, \( \varepsilon_b \), were evaluated according to ISO 527 Standard.\(^{23} \) In particular, \( E \) has been calculated by a least squares regression line fitted to the stress/strain curve in the range of values 0.0005 \( \leq \varepsilon \leq 0.0025 \). Three static tests were carried out for each specimen type.

After failure, the fracture paths were analyzed by using an AM4115ZT Dino-Lite digital microscope (with a magnification ranging from \( 20 \times \) to \( 220 \times \)) and fracture surfaces by means of a Scanning Electron Microscope Philips Quanta 400.

### 3 EXPERIMENTAL RESULTS

In the following, the experimental results in terms of fiber length distribution and tensile mechanical properties are presented.

As a preliminary finding, Figure 3 shows the FTIR spectra obtained for the vPPMF/GF and the rPPMF/GF compounds. In both cases, C–H stretching bands are present in the range 3000–2840 cm\(^{-1} \), while in the rPPMF/GF compound, the polyethylene (PE) peak centered at 719 cm\(^{-1} \) is visible, which is attributed to the methylene group rocking. The presence of PE in recycled PP is widespread, since they are popular polymers for use
in applications such as packaging, and therefore, the end of life material collection, separation, and recycling is critical.24

### 3.1 Fiber length distribution

The numeric and weighted mean fiber lengths were calculated from both the raw distribution (i.e., by using $N_i$ in Equations 4a and 4b) and the adjusted distribution (i.e., by using $N_{ad}$ in Equations 4a and 4b). All results are reported in Table 2, which shows that adjustment increases the weighted mean values by 4.1% up to 11.4%, where the highest increase was obtained for the distribution with the highest absolute value (vPPMF/GF_AM), since the picture sizes were the same for all analyses performed. On looking over the absolute results, it is seen that the recycled materials have always a lower mean length than the virgin ones, even though the same pellets were used for manufacturing the specimens. In particular, the rPPMF/GF.IM has $L_{w,ad}$ is equal to 352 $\mu$m against 421 $\mu$m of vPPMF/GF.IM, and for the AM materials, $L_{w,ad}$ is equal to 428 and 566 $\mu$m for recycled and virgin matrices, respectively. This outcome suggests that for a given manufacturing process and a set of process parameters, the postproduction fiber length of virgin and recycled specimens is likely influenced by the different rheological properties of the PP matrix. For the same matrix condition, the AM materials exhibit higher means values than the IM ones. More precisely, in the virgin condition, $L_{w,ad}$ of the AM samples is equal to 566 $\mu$m, that is, 34% higher than the IM ones. The same trend is valid for the recycled materials, where AM samples have $L_{w,ad}$ equal to 428 $\mu$m, which is 22% higher than the IM ones.

The adjusted distribution of the fiber length of each material is reported in Figure 4. The commonly observed right-skewed distribution’s shape of the fiber length can be described by the Burr Type XII distribution25,26:

$$F(L,a,b,c) = 1 - \left(1 + \left(\frac{L}{a}\right)^b\right)^{-c}; L > 0, a > 0, b > 0, c > 0$$  \hspace{1cm} (5)

$$f(L,a,b,c) = \frac{bc}{a} \left(\frac{L}{a}\right)^{-b-1} \left(1 + \left(\frac{L}{a}\right)^b\right)^{-c-1}; L > 0, a > 0, b > 0, c > 0$$  \hspace{1cm} (6)

where $F(L)$ and $f(L)$ are the cumulative distribution and the relevant probability density functions described by three positive parameters, namely, $a$, $b$, and $c$. The three Burr’s parameters were estimated by means of the maximum likelihood (ML) method implemented in MATLAB. The obtained ML estimates of all test series are reported in Table 2. The comparison between all estimated distribution functions is reported in Figure 5. Statistically shorter fibers can be observed for IM compared to AM materials and for recycled compared to virgin PP. Overall, by keeping in mind that the specimens were prepared starting from the same pellets, the most intense fiber breakage occurred during the fabrication of recycled injection-molded specimens. Focusing on the material influence on fiber breakage, it can be related to the different flow stresses of the PP caused by the different rheological properties of the recycled compared to the virgin matrices.27,28 It is worth mentioning that since the AM process parameters (see Table 1) were kept constant for all infill directions, the fiber length distribution has been assumed independent of the printing direction.

Characteristic fracture surfaces of virgin and recycled IM specimens were observed by SEM to evaluate qualitatively the through-the-thickness fiber orientation (FO). As expected in light of the relatively high specimens’ thickness ($t = 3.2$ and 3.4 mm, Figure 1), the characteristic skin–shell–core morphology was observed, as reported in Figures 6 and 7 for the virgin

| Material          | Number of measures | $L_m$ ($\mu$m) | $L_{m,ad}$ ($\mu$m) | $\Delta_m$ (%) | $L_w$ ($\mu$m) | $L_{w,ad}$ ($\mu$m) | $\Delta_w$ (%) | $a^c$   | $b^c$   | $c^c$ |
|-------------------|--------------------|----------------|---------------------|----------------|----------------|---------------------|----------------|---------|---------|-------|
| vPPMF/GF.IM       | 1063               | 306            | 327                 | 6.9            | 395           | 421                 | 6.6            | 729.7   | 2.22    | 5.26  |
| rPPMF/GF.IM       | 1301               | 285            | 297                 | 4.2            | 338           | 352                 | 4.1            | 423.5   | 3.07    | 2.77  |
| vPPMF/GF_AM       | 1399               | 396            | 435                 | 9.8            | 508           | 566                 | 11.4           | 539.3   | 2.70    | 1.97  |
| rPPMF/GF_AM       | 960                | 324            | 345                 | 6.5            | 402           | 428                 | 6.5            | 524.9   | 2.70    | 2.94  |

$\Delta_m = (L_{m,ad} - L_m)/L_m$.

$\Delta_w = (L_{w,ad} - L_w)/L_w$.

*Equations (5) and (6).*
and recycled IM specimens, respectively. In particular, Figures 6A and 7A show that the thickness of the core layer (highlighted by the yellow box) is approximately 1/2 of the specimen’s thickness, while Figures 6B and 7B and Figures 6C and 7C report details of the shell and core zones, respectively.

3.2 | Tensile test results

Figure 8 shows the engineering stress versus engineering strain curves obtained from the static tensile tests. In particular, Figure 8B,C reports the comparison between IM and AM specimens, for virgin and recycled materials,
respectively, while Figure 8A,D–F shows the influence of recycling for a given manufacturing process. The average values of $E$, $\sigma_m$, and $\varepsilon_b$ are listed in Table 3 and graphically reported in Figure 9 with the relevant standard deviation (s.d.). By considering the influence of recycling on IM specimens, the outcome is that all material properties degrade by 11.1%, 34.1%, and 26.5% for $E$, $\sigma_m$, and $\varepsilon_b$, respectively. A higher reduction of the elastic modulus equal to 20% was found for PP filled with MF (i.e., without GF) in a previous investigation\(^29\); conversely, a lower reduction of $\sigma_m$ was found previously (18.9%)\(^29\) compared to the present paper (34.1%). A tentative explanation regarding $\sigma_m$ will be given in Section 4.2.

Concerning the influence of recycling on AM specimens, significant deterioration of all analyzed properties can be noticed in Figure 9, which are markedly higher for two infill patterns, namely, $0^\circ$ and $90^\circ$. The reductions of mechanical properties are as follows: $E$ decreases by 32.6% and 39.7%, $\sigma_m$ decreases by 42.9% and 48.9%, and $\varepsilon_b$ decreases by 19.0% and 23.7% for $0^\circ$ and $90^\circ$ infill patterns, respectively. In the case of the infill pattern $\pm 45^\circ$, lower reductions were found, in that $E$, $\sigma_m$, and $\varepsilon_b$ of recycled materials are 12.9%, 19.6%, and 13.3% lower than the virgin ones, respectively (Table 3).

Regarding the influence of the manufacturing process on the material properties, Figure 8B shows that, although $v_{AM_F0}/C14$ specimens have an elastic modulus comparable to the $v_{IM}$ samples, they have lower $\sigma_m$ and $\varepsilon_b$ values by 15.7% and 53.6%, respectively. The stress–strain curves of $v_{AM_F0}$ are superimposed to those relevant to $v_{IM}$ specimens, and they can be considered as prematurely interrupted. This result will be justified later in light of the damage mechanisms reported in Sections 4.1 and 4.2. Differently, the stress–strain curves of $r_{IM}$ specimens are above those of $r_{AM_F0}$ samples, as reported in Figure 8C; however, a significant reduction of $\sigma_m$ and $\varepsilon_b$ for AM specimens still occurs, similar to Figure 8B.

Finally, concerning the influence of the infill pattern, Figure 8B,C shows that, as expected, the mechanical
The mechanical properties of the 3D printed materials are strictly dependent on the raster angle, and this holds true for both virgin (Figure 8B) and recycled (Figure 8C) materials. The infill pattern dependency of the mechanical properties seems correlated to the synergic effects of the material and manufacturing process adopted. Several investigations show that the mechanical properties of net polymers such as ABS and PLA are not significantly influenced by the infill direction. However, in the case of FDM SFRPCs, the mutual effects of voids and bulk anisotropy induced by the reinforcement lead to an infill pattern dependence of the mechanical properties. In the present investigation and according to the literature, best performance in terms of $E$ and $\sigma_m$ was obtained for raster angle 0°, while the 90° infill pattern produced the lowest properties with a percentage deterioration around 60%. The virgin and recycled AM_F±45° materials show the highest $\epsilon_b$, in agreement with the literature, and approximately three times higher than for the respective AM_F0° materials.

4 | FAILURE MODE AND DAMAGE ANALYSIS

The fracture surfaces of the broken specimens were analyzed at the macroscopic and microscopic level, by using the digital microscope and the scanning electron microscope, respectively. Observed damage mechanisms were related to material and manufacturing processes as reported in the following sections.

4.1 | Macroscopic analysis

Injection-molded virgin and recycled specimens exhibited similar macroscopic failure modes, as reported in Figure 10, with fracture surfaces almost perpendicular to the loading direction. This result is consistent with the literature regarding SFRPCs with a mold-fill direction parallel to the specimen's axis (see, e.g., Friedrich and Karger-Kocsis, Greenhalgh, and
| Material | Technology | Material condition | Specimen orientation | Infill pattern | $E$ (mean) (MPa) | $E$ (s.d.) (MPa) | $\sigma_m$ (mean) (MPa) | $\sigma_m$ (s.d.) (MPa) | $\epsilon_b$ (mean) (m/m) | $\epsilon_b$ (s.d.) (m/m) |
|---------|------------|-------------------|----------------------|---------------|----------------|----------------|---------------------|---------------------|---------------------|---------------------|
| v.IM    | IM         | Virgin            | —                    | —              | 7892           | 124            | 86.0                | 0.9                 | 0.0306              | 0.0009              |
| r.IM    | Recycled   | —                 | —                    | —              | 7014           | 38             | 56.7                | 0.8                 | 0.0225              | 0.0017              |
| v_AM_F0°| AM         | Virgin            | F                    | 0°             | 8036           | 386            | 72.5                | 5.7                 | 0.0142              | 0.0011              |
| v_AM_F90°| —         | 90°               | —                    | —              | 3518           | 273            | 28.4                | 3.1                 | 0.0232              | 0.0044              |
| v_AM_F±45°| —        | ±45°              | —                    | —              | 2981           | 415            | 29.6                | 1.6                 | 0.0331              | 0.0041              |
| r_AM_F0°| Recycled   | —                 | 0°                   | —              | 5414           | 82             | 41.4                | 0.7                 | 0.0115              | 0.0007              |
| r_AM_F90°| —         | 90°               | —                    | —              | 2120           | 65             | 14.5                | 0.4                 | 0.0177              | 0.0026              |
| r_AM_F±45°| —        | ±45°              | —                    | —              | 2595           | 48             | 23.9                | 2.7                 | 0.0287              | 0.0024              |
Hayes et al.\textsuperscript{41} and the references quoted therein). In the case of v\_AM\_F0\textdegree and r\_AM\_F0\textdegree specimens, the failure initiated at the start point of the fillet due to the presence of voids at the interface between the wall and the infill, generated by the peculiar infill strategy illustrated in Figure 2C and then propagated in a zigzag appearance (more pronounced for virgin than for recycled specimens), due to interlayer delamination and debonding of adjacent filaments.\textsuperscript{17} Corrugated macroscopic fracture surfaces almost perpendicular to the loading direction were observed in the case of v\_AM\_F\pm 45\textdegree and r\_AM\_F\pm 45\textdegree specimens, as shown in Figure 10. Finally, in the case of v\_AM\_F90\textdegree and r\_AM\_F90\textdegree samples, the fracture surface was planar, normal to the loading direction and located at the interface between adjacent filaments.

4.2 Microscopic analysis

Concerning the observations at the microscopic level, the first row of Figure 11 shows the characteristic fracture surfaces of virgin and recycled injection-molded specimens. In both cases, diffuse fiber pullout, broken fibers, and the presence of MF particles can be seen. Moreover, unlike v\_IM, in the case of r\_IM specimens, diffuse fiber–matrix debonding was observed, which probably induced cracks in the recycled matrix.\textsuperscript{42} as shown in Figure 12A,B: in fact, PP is highly susceptible to the oxidative degradation induced by the recycling process, which reduces the strength of the fiber–matrix adhesion.\textsuperscript{43} The diffuse fiber–matrix debonding is probably responsible for the decrease of $\sigma_m$ compared with the respective virgin material and for the higher decrease.
observed in the present MF filled plus GF-reinforced PP than in the previous MF filled (without GF reinforcement) PP.\textsuperscript{29}

Fiber fracture and fiber pullout were the typical damage mechanisms in the case of virgin and recycled AM\textsubscript{F0°} specimens, as shown in the second row of Figure 11. Moreover, the presence of voids can be clearly appreciated, which are larger in the case of recycled than virgin material, as well as of delaminations between adjacent layers, as reported by Rahimizadeh et al.\textsuperscript{17} A peculiarity of the fracture surfaces of virgin and recycled AM\textsubscript{F0°} specimens deserves the detailed view of Figure 13, which highlights the voids at the start point of the fillet caused by the “U-turn” of the deposition path necessary to infill the fillet areas (see Figure 2C). Furthermore, this infill strategy causes the filaments, and thus the fibers, to be unfavorably oriented locally with respect to the applied stresses. On this basis, the premature failures deduced by comparing the AM\textsubscript{F0°} stress–strain curves

| AM\textsubscript{F0°} | Delamination | Infill-based voids |
|----------------------|--------------|--------------------|

| AM\textsubscript{F45°} | Delamination | Layer thickness |
|-----------------------|--------------|-----------------|

\textbf{FIGURE 11} SEM analyses of fracture surfaces for the analyzed materials and manufacturing processes
A limited number of broken fibers were observed in the case of virgin and recycled AM_F90° specimens, as reported in the third row of Figure 11. In fact, the previous analysis at the macroscopic level has shown that the crack propagated at the interface between adjacent filaments, the delaminations between layers being practically absent. In more detail, diffuse fibrillation was observed in the case of virgin compared to recycled material, which implies that the virgin matrix underwent higher plastic deformation. Finally, considering the significant number of voids present in the wall and the lower elongation at break of AM_F0° compared to AM_F90° specimens (see Figure 8B,C), it may be argued that the failure initiated in the wall and then propagated in the infill.

Finally, diffuse interlayer delamination and debonding of adjacent filaments were observed in the case of AM_F±45° virgin and recycled specimens, as reported in the last row of Figure 11.

5 | CONCLUSIONS

The static tensile behavior of virgin and recycled short GF-reinforced PP filled with MF was investigated, by taking into consideration the influence of two manufacturing processes, namely, the IM and the pellet AM. In particular, IM specimens were produced with the injection flow direction parallel to the specimens’ longitudinal axis, whereas AM specimens were produced in the flat (F) position, by adopting three different infill patterns,
Recycled PP matrix degrades the mechanical properties in terms of elastic modulus $E$, tensile strength $\sigma_m$, and elongation at break $\varepsilon_b$, regardless of the manufacturing process. In the case of AM materials, degradation is more limited for the $\pm 45^\circ$ than for the $0^\circ$ and $90^\circ$ infill patterns; overall, high anisotropy was found in the additive manufactured specimens, owing to the different infill patterns adopted, the main outcome of the static tensile tests being (i) the superior stiffness and strength of $0^\circ$ compared to $\pm 45^\circ$ and $90^\circ$ specimens and (ii) the superior ductility of $\pm 45^\circ$ compared to $0^\circ$ and $90^\circ$ specimens.

Regarding the manufacturing processes and considering only the results relevant to AM_F0$^\circ$ specimens, the elastic modulus of the virgin AM specimens is comparable to the respective IM specimens, while the elastic modulus of the recycled AM samples is lower than the respective IM ones. Both virgin and recycled AM specimens have lower tensile strength and elongation at break than the respective IM samples; this outcome seems to have been promoted by premature failure caused by the peculiar infill pattern at the start point of the fillet.

Virgin and recycled AM_F0$^\circ$ specimens exhibited a zigzag appearance of the fracture surface, due to interlayer delamination and debonding of adjacent filaments. Corrugated fracture surfaces almost perpendicular to the loading direction were observed for AM_F$\pm 45^\circ$ specimens. AM_F90$^\circ$ samples exhibited planar fracture surfaces, normal to the loading direction and caused by debonding of adjacent filaments; despite the similarity of the fracture surfaces at the macroscale, a more detailed microscopic analysis highlighted more intense plastic deformation in the virgin than in the recycled polymer matrix.

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### AUTHOR CONTRIBUTIONS

Daniele Rigon: Paper conceptualization, writing, testing, and data analysis. Mauro Ricotta: Paper conceptualization, writing, and data analysis. Giovanni Ardengo: Testing and data analysis. Giovanni Meneghetti: Paper conceptualization, writing, data analysis, and acquisition of funding.

### DATA AVAILABILITY STATEMENT

Data are available on request from the authors.

### NOMENCLATURE

- $E$: material elastic modulus (MPa)
- $f_{l,ad}$: relative density of fiber length ($L_i$)
- $L$: fiber length ($\mu$m)
- $L_i$: center of the $i$th bin ($\mu$m)
- $L_m$: numeric mean fiber length ($\mu$m)
- $L_{m,ad}$: adjusted numerical mean fiber length ($\mu$m)
- $L_w$: weighted mean fiber length ($\mu$m)
- $L_{w,ad}$: adjusted weighted mean fiber length ($\mu$m)
- $N_{l,ad} (N_{l,wit} + N_{l,int})$: adjusted number of fibers having length between $L_i - \Delta L/2$ and $L_i + \Delta L/2$
- $N_{l,int}$: number of fibers that intersect the contours of an image having a length between $L_i - \Delta L/2$ and $L_i + \Delta L/2$
- $N_{l,wit}$: number of fibers measured from an image having a length between $L_i - \Delta L/2$ and $L_i + \Delta L/2$
- $\Delta L$: width fiber length bins ($\mu$m)
- $\varepsilon_b$: elongation at break (m/m)
- $\sigma_m$: tensile strength (MPa)

### REFERENCES

1. Dizon JRC, Espera AH, Chen Q, Advincula RC. Mechanical characterization of 3D-printed polymers. Addit Manuf. 2018;20: 44-67.
2. Cruz Sanchez FA, Boudaoud H, Camargo M, Pearce JM. Plastic recycling in additive manufacturing: a systematic literature review and opportunities for the circular economy. J Clean Prod. 2020;264:121602.
3. Godina R, Ribeiro I, Matos F, Ferreira BT, Carvalho H, Peças P. Impact assessment of additive manufacturing on sustainable business models in Industry 4.0 context. Sustain For. 2020;12:7066.
4. Hart KR, Frketic JB, Brown JR. Recycling meal-ready-to-eat (MRE) pouches into polymer filament for material extrusion additive manufacturing. Addit Manuf. 2018;21:536-543.
5. Czyżewski P, Bieliński M, Sykutera D, et al. Secondary use of ABS co-polymer recyclates for the manufacture of structural elements using the FFF technology. Rapid Prototyp J. 2018; 24(9):1447-1454.

6. Mohammed MI, Wilson D, Gomez-Kervin E, Tang B, Wang J. Investigation of closed-loop manufacturing with acrylonitrile butadiene styrene over multiple generations using additive manufacturing. ACS Sustain Chem Eng. 2019;7(16):13955-13969.

7. Vidakis N, Petousis M, Maniadi A, Koudoumas E, Vairis A, Kechagias J. Sustainable additive manufacturing: mechanical response of acrylonitrile-butadiene-styrene over multiple recycling processes. Sustain For. 2020;12(9):3568.

8. Pinho AC, Amaro AM, Piedade AP. 3D printing goes greener: study of the properties of post-consumer recycled polymers for the manufacturing of engineering components. Waste Manag. 2020;118:426-434.

9. Reich MJ, Woern AL, Tanikella NG, Pearce JM. Mechanical properties and applications of recycled polycarbonate particle material extrusion-based additive manufacturing. Materials (Basel). 2019;12:1642.

10. Pepi M, Zander N, Gillan M. Towards expeditionary battlefield manufacturing using recycled, reclaimed, and scrap materials. JOM. 2018;70(10):2359-2364.

11. Zander NE, Gillan M, Burckhard Z, Gardea F. Recycled polypropylene blends as novel 3D printing materials. Addit Manuf. 2019;25:122-130.

12. Zander NE, Gillan M, Lambeth RH. Recycled polyethylene terephthalate as a new FFF feedstock material. Addit Manuf. 2018;21:174-182.

13. Anderson I. Mechanical properties of specimens 3D printed with virgin and recycled polylactic acid. 3D Print Addit Manuf. 2017;4:110-115.

14. Cruz Sanchez FA, Boudaoud H, Hoppe S, Camargo M. Polymer recycling in an open-source additive manufacturing context: mechanical issues. Addit Manuf. 2017;17:87-105.

15. Zhao P, Rao C, Gu F, Sharmin N, Fu J. Close-looped recycling of polylactic acid used in 3D printing: an experimental investigation and life cycle assessment. J Clean Prod. 2018;197:1046-1055.

16. Tian X, Liu T, Wang Q, Dilmurat A, Li D, Ziegmann G. Recycling and remanufacturing of 3D printed continuous carbon fiber reinforced PLA composites. J Clean Prod. 2017;142:1609-1618.

17. Rahimizadeh A, Kaljian J, Fayazbakhsh K, Lessard L. Recycling of fiberglass wind turbine blades into reinforced filament for use in Additive Manufacturing. Compos Part B Eng. 2019;175:107101.

18. Stoof D, Pickering K. Sustainable composite fused deposition modelling filament using recycled pre-consumer polypropylene. Compos Part B Eng. 2018;135:110-118.

19. Veer FA, Setaki F, Riemsag AC, Sakkas P. The strength and ductility of glass fibre reinforced 3D-printed polypropylene. Heron. 2017;62:85-97.

20. ASTM D638-14. Standard Test Method for Tensile Properties of Plastics. West Conshohocken, PA: ASTM International; 2014.

21. Spoerk M, Holzer C, Gonzalez-Gutierrez J. Material extrusion-based additive manufacturing of polypropylene: a review on how to improve dimensional inaccuracy and warpage. J Appl Polym Sci. 2020;137(12):48545.

22. Fu S-Y, Mai Y-W, Ching EC-Y, Li RKY. Correction of the measurement of fiber length of short fiber reinforced thermoplastics. Compos Part a Appl Sci Manuf. 2002;33(11):1549-1555.

23. ISO 527-2019. Plastics—Determination of Tensile Properties. Standard International Organization for Standardization; 2019.

24. Larsen AG, Olafsen K, Alrock B. Determining the PE fraction in recycled PP. Polym Test. 2021;96:107058.

25. Burr IW. Cumulative frequency functions. Ann Math Stat. 1942;13(2):215-232.

26. Tadikamalla PR. A look at the Burr and related distributions. Int Stat Rev/Rev Int Stat. 1980;48(3):337-344.

27. Gupta VB, Mittal RK, Sharma PK, Mennig G, Wolters J. Some studies on glass fiber-reinforced polypropylene. Part I: reduction in fiber length during processing. Polym Compos. 1989;10(1):8-15.

28. von Turkovich R, Erwin L. Fiber fracture in reinforced thermoplastic processing. Polym Eng Sci. 1983;23(13):743-749.

29. Meneghetti G, Ricotta M, Sanità M, Refosco D, Atzori B. Notch sensitivity on fully reversed axial fatigue behaviour of different polylactide compounds. Procedia Engineering. 2015;109:441-449.

30. Ahmed AA, Susmel L. A material length scale-based methodology to assess static strength of notched additively manufactured polylactide (PLA). Fatigue Fract Eng Mater Struct. 2018;41:2071-2098.

31. Ng CT, Susmel L. Notch static strength of additively manufactured acrylonitrile butadiene styrene (ABS). Addit Manuf. 2020;34:101212.

32. Allum J, Moetazedian A, Gleadall A, Silberschmidt VV. Interlayer bonding has bulk-material strength in extrusion additive manufacturing: new understanding of anisotropy. Addit Manuf. 2020;34:101297.

33. Rigon D, Ricotta M, Meneghetti G. A literature survey on structural integrity of 3D printed virgin and recycled ABS and PP compounds. Procedia Struct Integr. 2020;28:1655-1663.

34. Ning F, Cong W, Qiu J, Wei J, Wang S. Additive manufacturing of carbon fiber reinforced thermoplastic composites using fused deposition modeling. Compos Part B Eng. 2015;80:369-378.

35. Spoerk M, Savandaiah C, Arbeiter F, et al. Anisotropic properties of oriented short carbon fibre filled polypropylene parts fabricated by extrusion-based additive manufacturing. Compos Part a Appl Sci Manuf. 2018;113:95-104.

36. Peng X, Zhang M, Guo Z, Sang L, Hou W. Investigation of processing parameters on tensile performance for FDM-printed carbon fiber reinforced polylactide 6 composites. Compos Commun. 2020;22:100478.

37. El Magri A, El Mabrouk K, Vaudreuil S, Ebn Touhami M. Mechanical properties of CF-reinforced PLA parts manufactured by fused deposition modeling. J Thermoplast Compos Mater. 2021;34(5):581-595.

38. Somireddy M, Singh CV, Czekanski A. Mechanical behaviour of 3D printed composite parts with short carbon fiber reinforcements. Eng Fail Anal. 2020;107:104232.

39. Friedrich K, Karger-Kocsis J. Fractography and failure mechanisms of unfilled and short fiber reinforced semi-crystalline thermoplastics. In: Roulis-Moloney AC,
40. Greenhalgh ES. *Failure Analysis and Fractography of Polymer Composites*. CRC Press, Woodhead Publishing Limited; 2009.

41. Hayes MD, Edwards DB, Shah AR. *Fractography in Failure Analysis of Polymers*. PDL Handbook Series. William Andrew Publishing; 2015.

42. Huang H, Talreja R. Numerical simulation of matrix micro-cracking in short fiber reinforced polymer composites: initiation and propagation. *Compos Sci Technol.* 2006;66(15):2743-2757.

43. Yang L, Thomason JL, Zhu W. The influence of thermo-oxidative degradation on the measured interface strength of glass fibre-polypropylene. *Compos Part a Appl Sci Manuf*. 2011;42(10):1293-1300.

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