

# Recycling of Plastic Waste by Integrating Extrusion and Additive Manufacturing Techniques: Lessons Learned from RELIVE Project

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**Abstract.** Polymer processing in modern industry is often not efficient from the point of view of energy and material consumption. This production system must be revised to pursue a circularity of products and materials. Recent developments in additive manufacturing technologies for polymers and polymer-based composites are enabling changes of production paradigm from “Design for Manufacturing” to “Manufacturing for Design”, providing new intriguing lightweight solutions. This paper summarizes the lesson learned from the RELIVE project “REcycling of pLastic wastes integrating extrusion and additIVE manufacturing techniques”, in terms of methodology and results.

## Introduction

The environmental challenges are global and systematic. To achieve the long-term sustainable targets determined by the European Union, the core system of society needs to be dramatically modified. The production paradigm in modern industry is linear and oriented to the consolidated extraction-transformation-usage-disposal chain. This way of thinking for the production system is not feasible any longer and must be changed. In recent years, the development of innovative processes set new approaches in production, introducing the concept of “Manufacturing for Design” as an alternative to the consolidated “Design for Manufacturing”. Modern manufacturing is often not efficient from the point of view of energy consumption and efforts to reduce environmental impact by lifecycle management initiatives in manufacturing have been presented recently [1]. In this framework, process digitalization and automation are increasingly adopted to improve resource efficiency, as widely explored in advanced composite manufacturing routes where defect reduction and automated programming strategies are pursued to minimize material waste and rework [2,3]. In the traditional economic system, materials are mostly extracted from fossil sources and the products, obtained from the transformation of raw materials by consuming energy, at the end of their life cycle, are thrown away as waste. This production system must be stopped to pursue a circularity of products and materials [4]. Circular economy and sustainable manufacturing represent promising strategies to reduce the impact of human activities on climate changes. Nowadays, the mainstream for advanced and mass production sectors must be the development of new strategies based on total life-cycle approaches and on the 6R concept (Reduce-Reuse-Redesign-Remanufacture-Recycle-Recover). In particular, lifecycle-based metrics are becoming central also for lightweight structures, as shown by recent LCA studies on composite components for e-mobility applications [5], reinforcing the need for production strategies that couple performance with measurable sustainability benefits. As far as polymeric materials are concerned, the continuous growing market and usage in different fields

together with the constant increase of their waste highlight the relevance of a proper recycling strategy both for economic and social reasons. Furthermore, the residual value of this waste has been stimulating the development of different recycling routes based on mechanical, thermal or chemical approaches to maximize recoverable energy in more sustainable and economically effective perspectives. Currently, the plastic economy is highly linear, but the transition to a circular economy, retaining plastics at their highest value [6], is always more essential considering their massive consumption and the adverse impacts on the environment if their wastes are not processed properly [7]. In the past decades, several industries assumed lightweight polymer-based materials as a driving trend at the design stage [8]. The wider usage of polymer-based materials already provides some environmental benefits considering their enhanced manufacturability and lightness. Nevertheless, the concept itself of producing polymers, often not renewable materials on an acceptable time scale, releasing them again in the environment as wastes, is far from any idea of sustainability [9,10]. In this historical period, the implementation of agile integrated manufacturing approaches based on the principle of reusing waste is mandatory to reduce the environmental impact in production [11].

Fused filament fabrication (FFF) technology has been addressed as a key manufacturing approach able to reduce the consumption of material [12]. FFF is a 3D printing process that allows parts production by a “layer-by-layer” approach and, nowadays, it is a consolidated method exploited to manufacture complex and performant end-use products. Additive manufacturing in general has been defined by the European Community as an advanced technology that can “offer a range of opportunities that will enable European industry to expand its leadership in the emerging markets for the products and services of the future” [13]. Fused filament fabrication (FFF) is a consolidated technology within the panorama of additive manufacturing technologies, but it is mostly employed to process near-amorphous low-performance thermoplastics. Recently, the introduction of reinforcing (long or short) fibers inside the filament has further spread the usage of FFF to obtain advanced products. Reusing end-of-life thermoplastics for making filaments employed in FFF can be a relevant step to circular economy production. This manufacturing strategy is not rigorously a new idea [14]. Nevertheless, the recovery and treatment of these wastes often imply a reduction in mechanical and physical properties [15]. Consequently, regenerated filaments are poorly appealing and are marginally used only for secondary production. Therefore, the property control of parts produced from recycled or reprocessed thermoplastics is strictly required [16]. Enhancing the polymeric properties by inclusion of opportune fillers and reinforcing fibers can be pursued in filament extrusion, also to counter warp and shrink phenomena, which the processed semi-crystalline polymers can be subjected to [17]. Recent experimental evidence demonstrated the potential of recycled plastics such as polypropylene and polyethylene terephthalate as new FFF feedstock materials. Anyway, the absence of in-depth process-property correlations as well as the scarce availability of theoretical models lead to insecurity of potential customers [18]. Similar needs for robust process understanding, traceability and defect mitigation are well-known in multiple composite manufacturing processes, where impregnation/permeability phenomena and the control of preform processing conditions are key to part quality [19,20]. In addition, also energy-efficient solutions have been investigated to enhance process control and potentially reduce cycle times and energy consumption [21,22]. In literature, some results concerning the temperature and velocity fields of the thermoplastics and their relationship with the formation of the beads and the layering of the extruded materials are reported [23]. Other researchers conducted simulations aiming to assess the influence of the nozzle geometries on the flow behavior of the thermoplastics [24]. Strict assumptions have been accepted in the development of these models, which involve for example the simplification of the interaction between the fibers, the hypothesis of very low fiber volume fraction or of a Newtonian fluid behavior for the thermoplastic melt [25]. New models and tools to predict flow, fiber orientation, neck and bond formation and chain diffusion, for instance, are desirable, assuming early approaches, derived from sintering and thermoplastics welding [25] and accounting for transient thermal and molecular diffusion [26]. Similarly, the cooling down of the deposited filament and its solidification are critical factors influencing the products’ shape and stability [27].

The RELIVE project, “REcycling of pLastic wastes integrating extrusion and additIVe manufacturing techniques”, funded by the Italian Ministry of University and Research under the PRIN program (Prot. N. 2022FBB37P), explored the feasibility of integrating additive manufacturing of reinforced polymers by FFF with the mechanical recycling of thermoplastic wastes, proposing a direct strategy able to bring plastic waste to new life and addressing major economic and technological restraints. More specifically, shredded thermoplastic waste was characterized according to material typology, purity and processability by thermal and rheological analyses. Subsequently, the ground materials were extruded, as they are or with the addition of reinforcing fibers, into wires to be fed directly to a nozzle for the final transformation using the FFF technique. Experimental analysis, numerical modelling and simulation activities were conducted to elucidate advantages and limits of the proposed solution. The implementation of the RELIVE project has been articulated in five work packages (WPs), as depicted in Figure 1. The first (WP0) and one (WP4) have been devoted to management and dissemination initiatives. Technical WPs, namely WP1, WP2, and WP3, aimed to:

- Identification, at least at a qualitative level, of the composition of the secondary raw materials through thermal and molecular investigations;
- Homogenization of recycled plastics by extrusion under suitable conditions and analysis of the rheological behavior of extruded materials;
- Systematic investigation of the extruded filaments and subsequent deposition by FFF using innovative tooling and machines;
- Assessment of the influence of the integrated process parameters on the ultimate performance of additively manufactured products;
- Numerical analysis and optimization of the proposed manufacturing route looking also at the efficiency of the proposed recycling strategy.

This paper summarizes lessons learned from the RELIVE project in terms of methodology and main results. More details could be found in focused papers published by the authors elsewhere.

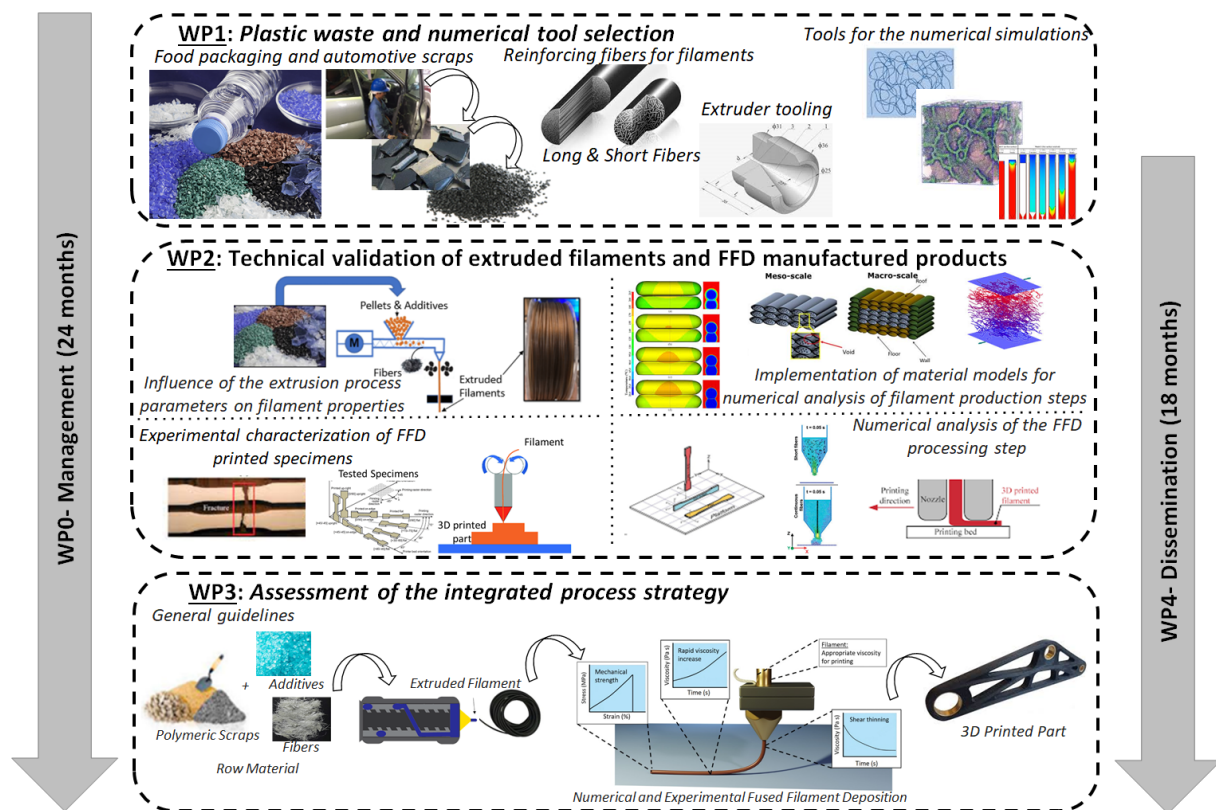


Fig. 1. Schematic of the RELIVE project.

## Materials and Methods

### Feedstock selection

The rationale behind the selection of waste polymeric materials to be recycled as a reinforced filament for additive manufacturing by FFF was based on materials processability by extrusion and on potential industrial applications. Given the considerable variety of polymers used for industrial applications and, therefore, of potentially acquirable waste for this research, special interest has been addressed to two mass sectors: food packaging and automotive. In the first area, given the relatively short average lifespan of packaging and the intrinsic properties of the polymeric materials typically used, polyolefins and increasingly bio-based plastics, the recycling strategy explored in this research could alleviate the environmental impacts associated with both waste and the end-of-life of these products, which are still largely disposed of in landfills. The second sector addressed, the automotive industry, in addition to using significant quantities of polypropylene for various components, accounts for nearly 40% of the total demand for polyamide-based engineering plastics (PA). Here too, despite the greater durability of the components, recycling, both industrially and for end-of-life products, remains challenging, given that these polymers can undergo thermomechanical degradation if not properly reprocessed, with ultimate performance further impaired by the aging processes inevitably experienced by the components during their life. That said, the research focused on polylactic acid (PLA) and polyamide 6 (PA6) mechanically recycled by extrusion and reprocessed to obtain filaments, even filled with long glass fibers, using FFF technology. In detail, filament production was performed using an innovative rotating impregnation die, previously patented by some of the authors [29], specifically developed for the manufacturing of fiber-reinforced polymer filaments suitable for FFF. The die enhances the interaction between the polymer matrix and the fibers. By inducing a swirling flow within the melt, the rotation improves fiber wetting and promotes partial fiber interlacing. The custom impregnation die consists of a static section coupled to the extruder, which supplies the polymer melt. Following a 90° redirection, the melt stream meets the reinforcing fibres, which are impregnated and guided coaxially toward the die exit. The fibres are fed from the top and preheated via an auxiliary heating band; this prevents thermal gradients in the polymer and inhibits backflow. A thermocouple is employed to monitor and regulate the preheating temperature. Subsequently, the polymer-fibre bundle enters a rotating die section that induces a swirling flow to enhance fibre wetting. This rotation is driven by a 24 V gear motor through a chain-and-pinion transmission. Before discharge, a fixed nozzle suppresses the swirling motion and realigns the flow, ensuring thorough impregnation and dimensional stability of the final composite filament. The subsequent stage involves the deposition of the composite filaments utilizing a custom FFF system. The material is fed into an extrusion head and deposited onto a heated build plate maintained at 55 °C. The composite filament is driven through a 1.2 mm nozzle and extruded in the molten state at a temperature of 180 °C. Both the co-extrusion and deposition processes, including the resulting filament outputs, are illustrated in Figure 2. The recycled materials were systematically characterized and analyzed with experimental tests and theoretical analyses.

### Experimental tests

Recycled extruded materials were characterized in terms of thermal properties and rheological behavior, while the filaments, including those reinforced with long glass fibers were analyzed through quasi-static tensile tests and morphological-structural aspects. Calorimetric measurements were carried out using a TA Instruments DSC Q2000 equipped with a refrigerated cooling system (RCS) and a nitrogen purge flow of 50 mL/min [30]. The standard protocol consisted of a first heating scan from room temperature (RT) to 200 °C to erase the previous thermomechanical history, followed by a short isothermal step (1 min) at 200 °C, a controlled cooling from 200 °C to RT, and a second heating scan over the same temperature range to identify the intrinsic thermal transitions of the material. In addition, a dedicated set of experiments was performed by varying the cooling rate during the cooling step in order to assess the material response under both uniform and non-uniform cooling conditions, and to evaluate the influence of cooling history on the subsequent thermal behavior.

Melt rheological behavior was investigated under controlled thermal and atmospheric conditions by means of rotational rheometry using a modular compact rheometer (MCR 302, Anton Paar GmbH). Small-amplitude oscillatory shear measurements were used to determine the complex viscosity, and the data were converted into an equivalent shear viscosity through the Cox–Merz rule, allowing the reconstruction of the dynamic viscosity surface  $\eta(T, \dot{\gamma})$  over shear-rate and temperature ranges representative of filament manufacturing conditions. The resulting viscosity data were fitted using the Cross–WLF constitutive model, expressed as:

$$\eta(T, \dot{\gamma}) = \frac{\eta_0(T)}{1 + \left( \frac{\eta_0(T) \dot{\gamma}}{\tau^*} \right)^{1-n}} \quad (1)$$

where  $\eta(T, \dot{\gamma})$  is the shear viscosity (Pa·s),  $\dot{\gamma}$  is the shear rate ( $s^{-1}$ ), and  $\eta_0(T)$  is the zero-shear viscosity, which accounts for the temperature dependence of the melt viscosity. The parameters  $\tau^*$  (Pa) and  $n$  (–) are, respectively, a critical stress controlling the onset of shear-thinning and the power-law index describing the extent of shear-thinning ( $n < 1$ ). Furthermore, filaments were preliminary tested using an Instron model 5564 dynamometer on 8 cm-long specimens, pre-conditioned at room temperature and 50% relative humidity for 24 hours, setting the crosshead speed to 2 mm/min and a gauge length of 30 mm. Successive characterization was conducted at filament as well as sample level, by means of standard microscopic analysis, XCT analysis and mechanical testing. XCT scans were performed using uniTOM HR X-ray tomography system, operating at tube voltage of approximately 110 kV and current value of about 50  $\mu$ A, providing an isotropic voxel resolution of 6  $\mu$ m. Data processing, namely segmentation and distribution and orientation analysis, was performed in the MATLAB environment included, using a dedicated MATLAB script specifically developed to identify the regions of interest corresponding to the reinforcing fibers and the matrix. Afterward relevant microstructural descriptors (fiber volume fraction, spatial distribution, and voids) were calculated through a structure tensor–based approach. The resulting information provides insight into the quality of the filament production process and supports the interpretation of the mechanical behavior of additively manufactured composites based on recycled thermoplastic materials. Tensile and flexural characterizations were performed using a universal testing machine (MTS Criterion® Electromechanical Test System) equipped with a 1 kN load cell. For the tensile tests, ten specimens per filament type were evaluated at a constant crosshead speed of 1 mm/min. To prevent gripping damage, individual filaments were mounted on rigid supports prior to testing. The tensile modulus was derived from the linear slope of the stress–strain curves. Flexural tests were conducted on composite samples with a support span of 51 mm and a displacement rate of 1.35 mm/min. Tensile and flexural testing protocols followed the ISO 11566 and ASTM D790-15 standards, respectively.

### Numerical analysis

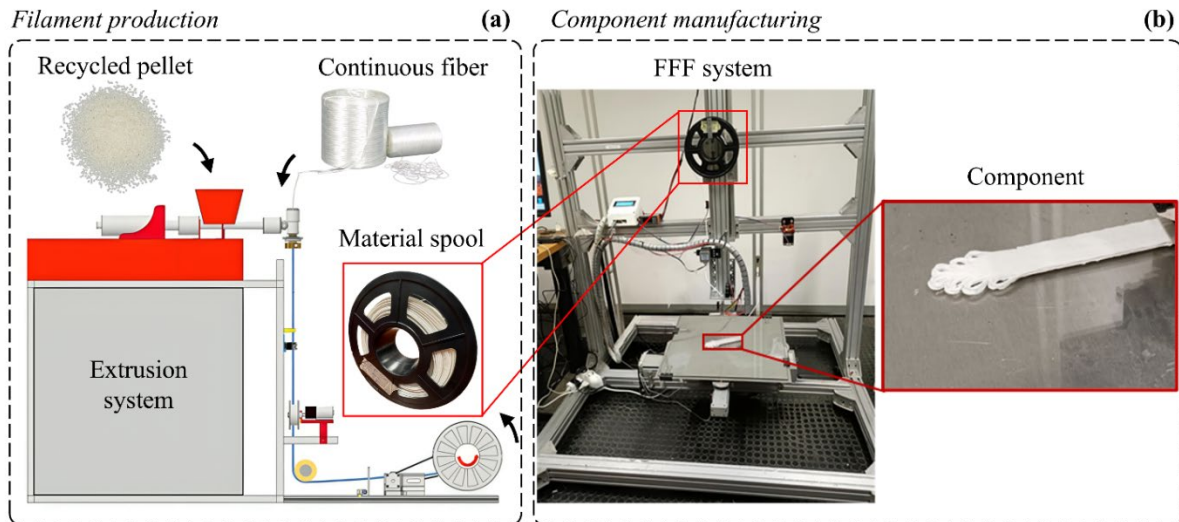
The deposition was investigated through transient CFD simulations under the assumptions of laminar, incompressible, non-isothermal flow of a highly viscous non-Newtonian polymer melt. The process was modelled, using the ANSYS-FLUENT package, as a two-phase system using a finite volume formulation, where the interface is captured by a Volume of Fluid (VOF) approach based on the transport of a volume-fraction field. This framework enables tracking the free surface evolution of the extrudate during its exit from the nozzle, contact with the substrate, and progressive cooling and, consequently, solidification. The governing physics included mass and momentum conservation coupled with the energy equation to account for heat transfer between the molten polymer, the surrounding air and the build plate. The melt rheology was introduced through a temperature and shear-rate dependent viscosity law, calibrated on experimental data as previously explained. In particular, the same Cross–WLF constitutive formulation used for the characterization was adopted to reproduce both shear-thinning behavior and the strong temperature sensitivity typical of recycled thermoplastics during extrusion and deposition. For numerical robustness and to represent the transition toward solid like behavior, viscosity bounds were imposed consistently with the experimentally observed processing window. In addition, phase change was accounted for through

an enthalpy-porosity formulation, enabling the progressive transition from melt to solid to be represented within the energy equation by introducing a liquid fraction field and a permeability like damping term in the momentum balance. The enthalpy-temperature relationship required by this approach, including the latent-heat contribution and the relevant transition range, was defined using the DSC data previously obtained, thereby ensuring that the onset and evolution of solidification in the simulations were consistent with the experimentally measured thermal behavior. The computational domain reproduced the nozzle-substrate interaction, allowing an efficient yet physically meaningful description of bead formation and thermal gradients. Boundary conditions were defined to mimic realistic manufacturing conditions, the nozzle walls were treated as no-slip boundaries kept at the extrusion temperature, whereas the substrate was modelled as a moving wall with prescribed speed and controlled temperature to reproduce deposition kinematics and heat extraction. Pressure outlet conditions were applied at the open boundaries to emulate an ambient environment. Additional modelling ingredients included surface tension at the polymer/air interface and thermal coupling among phases. The simulations provided useful qualitative and quantitative insights into the deposition mechanisms, highlighting the role of temperature driven viscosity evolution, interface stability, and cooling conditions on bead morphology. In the present CFD model, fiber effects were not explicitly included, in fact the melt was treated as an equivalent single-phase non-Newtonian matrix. Nevertheless, complementary modelling strategies are available for fiber-reinforced material extrusion. In particular, in a recent work a FEM based framework coupled with orientation tensors and an orientation-averaging homogenization scheme was used to incorporate process-induced anisotropy, stemming from mesostructure orientation, into component-scale predictions of deformation/warpage [28].

## Results and Discussion

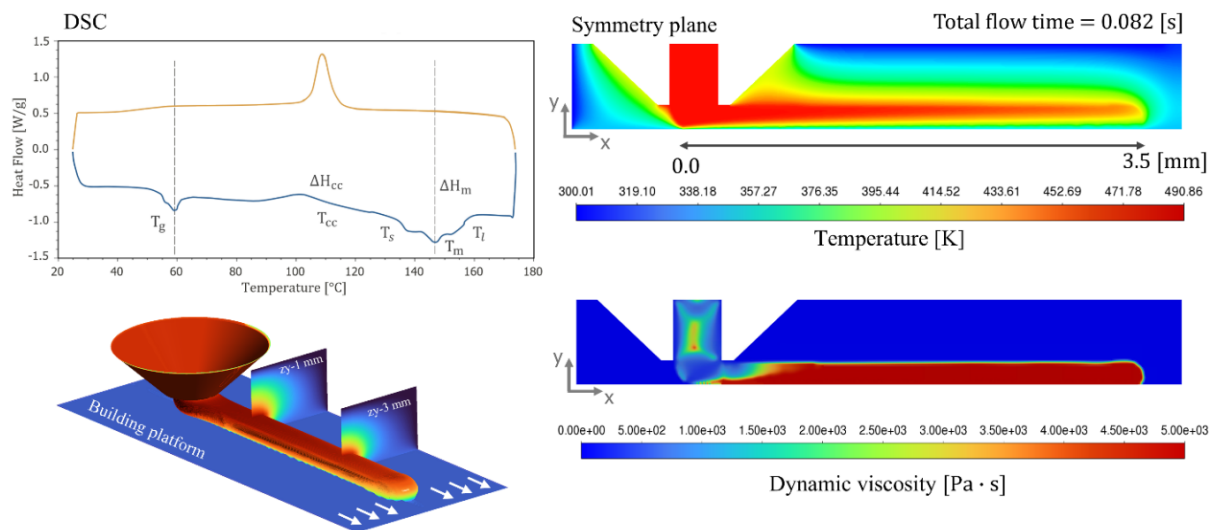
The RELIVE project explored the opportunities that can be derived by using plastic wastes to shape new products in a straight manufacturing route that starts from. In this case, the main technological challenges are represented by both the lower performance of plastic materials at the end of their life compared to raw materials, due to inevitable aging phenomena suffered by them during their useful life, and by the usual heterogeneity of considered wastes. The philosophy behind the proposed approach is quite simple and involves, after the section of the waste materials, the identification of the manufacturing process to be used for reshaping, the characterization of the materials properties, and the definition of strategies for properties enhancement. In the framework of the project, the investigated case study was given by the mechanical properties' enhancement coming from combination by co-extrusion of fibers and plastic waste integrated online with the additive manufacturing step. In Figure 2, the plastic waste, the recycled filaments and the deposited sample, processed during the RELIVE project, are shown.

The considered plastic wastes, namely PLA and PA6, were characterized by specific rheological and thermal properties, which essentially depend on their molecular and structural characteristics. It should be noted that in this investigation the starting material was already sorted, nevertheless it is not excluded to extend the approach to mixtures of different polymers by introducing a preliminary sorting operation. Thermo-mechanical characterization pointed out first the alteration of the properties of the materials and, consequently, of the processing window.



**Fig. 2.** (a) Schematic representation of the process; (b) Configuration of the customized FFF platform.

An integral part of the procedure was the redefinition of the processing window, replacing trial and error procedures and relying on appropriate numerical modelling and simulation. Reasonably, this procedure could be further improved in the future, using opportune AI tools, however, at the moment the trusted data available on recycled materials are not enough to build a robust database. In Figure 3, some results provided by the thermal characterization and the numerical simulation of the deposition are depicted.

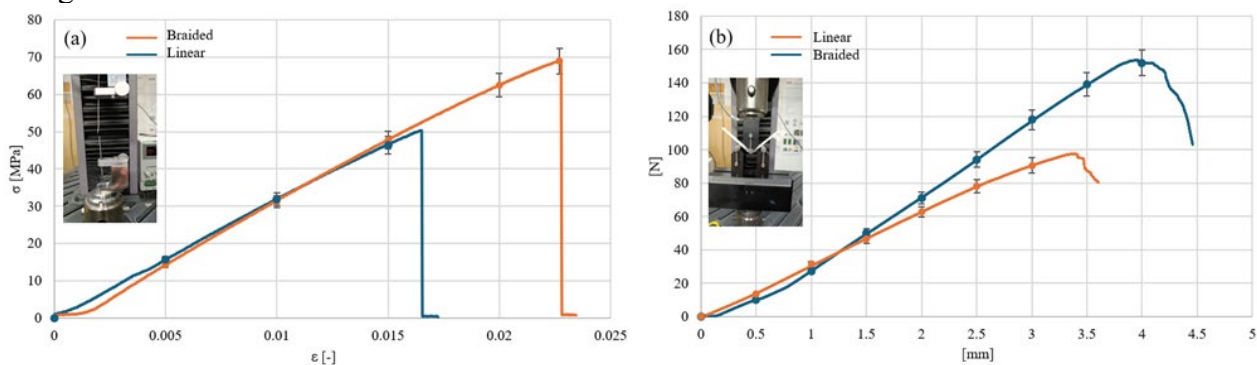


**Fig. 3.** DSC integrated data into temperature and viscosity fields evolution on the free surface and symmetry planes of the deposited bead.

Numerical simulations showed that the coupled thermal and rheological fields govern both flow stability inside the nozzle and shape retention after extrusion. In particular, the DSC curve in Figure 3 was used to define the melting-related temperature window relevant for extrusion, to extract the latent heat implemented in the enthalpy based formulation and to identify the solidus and liquidus temperatures adopted to model the phase change interval. Consistently with these thermal inputs, the predicted temperature distribution highlights a steep axial gradient along the nozzle and an even faster cooling of the extrudate immediately after deposition. As a direct consequence, the dynamic viscosity contour plots reveal a pronounced viscosity stratification: within the nozzle, high shear rates near the walls activate shear-thinning and maintain a low-viscosity “lubricating” layer, whereas the lower-shear core remains comparatively more viscous, thus giving insight on temperature process window

to optimize processability. Furthermore, during deposition, the rapid temperature decay causes a sharp increase in viscosity, progressively suppressing flow and enabling the filament to transition from a deformable melt to a stable bead. Overall, the simulations indicate that maintaining the melt above the phase change interval up to the nozzle exit is essential to ensure continuous extrusion, while a controlled post extrusion cooling rate is critical to achieve dimensional stability and reliable interlayer bonding for coalescence purpose.

Figure 4 provides a qualitative comparison of the representative results obtained from tensile and flexural testing of the filament specimens. Two distinct filament configurations, linear and braided, were evaluated under various loading conditions. As visible in Figure 4 (a), the tensile stress–strain curves of the glass fiber reinforced plastic (GFRP) filaments exhibit an initial linear elastic response for both configurations, followed by brittle fracture without significant plastic deformation, a behavior characteristic of fiber-reinforced polymer composites. The braided filaments achieve higher ultimate strain and tensile strength compared to the linear filaments, suggesting enhanced load transfer efficiency between the fibers and the matrix and improved resistance to localized defects. Conversely, the linear filaments fail at a lower strain, consistent with a highly aligned but less damage-tolerant structure. Specifically, the developed composites exhibited tensile strengths of approximately 50 MPa and 70 MPa for the linear and braided filaments, respectively. This represents an increase of roughly 60% and 115% compared to typical values for virgin PLA filaments of comparable diameter (ranging from 30 – 40 MPa). When compared with the literature on glass-fiber reinforced PLA, the braided filaments demonstrate excellent properties, matching the performance of optimized commercial composite filaments (typically ~60 MPa [31]) and reaching values comparable to recent scientific studies on glass-reinforced extrusion (~70 MPa [32]). The introduction of continuous fiber reinforcement effectively counteracted the thermomechanical degradation typical of the recycled matrix. This yielded stiffness values of 3.225 GPa and 3.256 GPa for linear and braided composites, respectively. A significant improvement over virgin PLA (0.813 GPa) aligning with the 3 – 3.5 GPa range reported for industrial composites [31, 32]. Regarding Fig. 4(b), the load–displacement curves from the three-point bending tests indicate that the braided configuration provides higher load-bearing capacity and flexural stiffness. These specimens sustain higher peak forces and demonstrate an extended quasi-linear regime, suggesting effective stress redistribution facilitated by the interlaced fiber architecture. In contrast, the linear specimens reach failure at lower displacements and peak loads, indicating a reduced capacity to delay crack initiation and propagation. The limited scatter in both tensile and flexural data (standard deviation of about 5%) confirms high repeatability and consistent manufacturing and testing conditions. Finally, the outcomes demonstrate that the innovative braiding technology improves both tensile and flexural performance, increasing strength and structural reliability. The observed differences can be attributed to the effect of fiber orientation, constraint of matrix voids, and the improvement of stress concentrations to the braided configuration.



**Fig. 4.** Representative data from (a) tensile and (b) flexural tests.

The idea behind the RELIVE project well reflected the concept of modeling the role of the circular economy for climate change mitigation, widely recalled in Horizon Europe Framework Programme (Horizon Europe) with the aims of developing novel, safe, environment friendly and commercially attractive methods of recycling a wide range of composite materials proposing tools that will enable

us to demonstrate the circularity and the environmental benefits of the solutions tested (HORIZON-CL4-2021-RESILIENCE-01). In the same direction, the HORIZON-CL4-2021-RESILIENCE-01-26 has been focused on enhancing efficiency and contributing to less waste and emissions while improving material/product/process quality all along the lifecycle of a service system. In a similar fashion, the project goals moved in the direction of contributing to decreasing dependency of Europe on imported raw materials.

## Conclusion

The RELIVE project explored the feasibility of integrating additive manufacturing of reinforced polymers by FFF with the mechanical recycling of thermoplastic wastes, proposing a direct strategy able to bring plastic waste to new life. The implemented activities and achieved outcomes confirmed the initial hypothesis concerning the reference paradigm. In particular, the definition of innovative “from waste-to-product” integrated additive approach, provides the following tangible impacts:

- extending the life and/or the applicability of polymeric materials even for high-end application by introducing specific additive and/or reinforcements (short and long fibers) in their recycling phase, overcoming the limits in the recycling of end life plastics derived by the inevitable degradation of the properties of the pristine materials;
- application of numerical approaches, eventually using advanced tools derived from the Industry 4.0 paradigm, in both the manufacturing and testing stages allows engineers to virtually test novel strategies reducing the usage of materials and resources.

Additionally, the investigated paradigm or approach reasonably provides a reduction of environmental burden in comparison with landfill and combustion, combined with greater value recovering from the thermoplastic wastes at the end-of-life of products. This aspect deserves further investigation and quantitative analysis using, for instance, LCA tools.

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