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Fused Deposition Technique for Continuous Fiber Reinforced Thermoplastic

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Abstract:

A simple technique for the production of continuous fiber-reinforced thermoplastic by fused deposition modelling, which involves a common 3D printer with quite limited modifications, is presented. An adequate setting of processing parameters and deposition path allows to obtain components with well enhanced mechanical characteristics compared to conventional 3D printed items. The most relevant problems related to the simultaneous feeding of fibers and polymer are discussed. The properties of obtained aramide fiber reinforced polylactic acid (PLA) in terms of impregnation quality and of mechanical response are measured.

1. Introduction

Polymer fused deposition technique, usually referred as fused deposition modelling (FDM) or, more commonly, 3D printing of plastic components, has found a dramatic commercial success thanks to its extremely broad application areas, to the flexibility of the process, to the versatility of materials that can be employed and, last but not least, to the low costs of printers. Single or multiple, integrated objects with good details definition and complex shapes can be produced from a drawing in fairly short times; low cost small series articles, prototypes and demonstration models can thus easily be obtained.

The large majority of such applications responds to aesthetical and/or functional needs linked to the shape or color of produced objects. Beside the use of modified material to get particularly appealing aesthetical effects, recent developments have been directed to the improvement of functional characteristics such as electrical/thermal conductivity, magnetic properties, optical response, etc. The

availability of such functionalized material gives the opportunity to put in practice a number of high level applications otherwise impossible [1-5].

On the other hand, mechanical and thermal performance constraints of the materials employed pose serious limitations to a further extension of this manufacturing technique to components with significant load carrying capacity in different environmental conditions. In relation to mechanical/structural performances, the polymeric materials usually employed, such as PLA, ABS, PC or Nylons present quite limited mechanical properties either coming from intrinsic low stiffness and strength of the polymer, either due to the defects which are invariably present in the processed materials such as voids, crystallinity variations, weld spots [6, 7].

The use of polymeric materials filled with reinforcing particles or short fibers can, to some extent, improve materials mechanical performances, still allowing the use of the same printers employed with unfilled materials; for these materials, however, issues related to a higher melt viscosity and short fibers orientation must be afforded, which affect the material proper formulation and the amount of reinforcing matter that can be added [8]. The addition of discontinuous fillers generally leads to increase of stiffness, while adverse effects over strength, brittleness and fatigue behavior are usually registered [8-10].

Certainly, the possibility of employing continuous reinforcing fibers, namely carbon, aramide or glass, may lead to products with much higher mechanical performances which are potentially useful for advanced applications. At present, quite few examples of polymer fused deposition processes for the production of continuous fiber composite items are found [11, 13]. The limited amount of reinforcing fiber, the wetting of deposited fiber bundles, the low deposition resolution and consequent presence of dry spot are only some of the difficulties which must be afforded in obtaining good quality products. Remarkable improvement in mechanical performances of deposited composites are claimed by Markforges, which employs a patent-pending method involving the extrusion of a continuous fiber reinforced composite filament; in this case, specific filaments in the form of "towpreg" are deposited with a specially designed printer [14].

In this paper, a simple modification of the extruding head of a commercial 3D printer to produce continuous aramide fiber reinforced PLA items is presented, in which dry fibers and thermoplastic polymer rod are simultaneous fed into a FDM extrusion die. The basic problems related to the feeding of continuous fibers to the extruded polymer melt, the geometrical limitations of obtainable objects and the chances of reinforcement orientation control are preliminary discussed. Considerations about the selection of optimal working parameters are presented in relation to the different object shapes and geometries tested at the moment. A quality evaluation is carried out of the final products and the presence of defects, such as surface irregularities, internal voids, fibers misalignments based on visual inspection, electron microscope observations, DSC analysis is assessed. Measurement of fiber content and mechanical performances in terms of strength and stiffness evidence the potential advantages of this technique for stiff and load carrying components.

2. Process issues

The simultaneous insertion of continuous fibers and solid polymer in the extrusion head of a FDM printer poses a number of difficulties which need to be afforded. The main issues are here discussed.

2.1 Fiber content and feed

To reach adequate improvement of mechanical properties in terms of stiffness and strength, deposited material should consist of a polymer matrix reinforced with a fair amount of continuous fibers. It should be considered that in the fiber direction the stiffness of the deposited material is proportional to the fiber content, in accordance to the mixture rule, $E_c = \Phi E_f + (1 - \Phi)E_m$, where E_c , E_f , E_m indicate the stiffness of final material (composite), fibers and polymer matrix respectively; Φ is the volume fraction of the fibers in the final material. With some approximations, the strength can also be estimated by a proportionality rule similar to the mixture rule previously indicated.

FDM printers extrusion heads usually consist of circular dies with diameters as low as .7 to 1 mm; such dimensions allow to deposit very small drops of polymer thus reaching limited surface roughness and accurate shape reproduction. Since the solid polymer rod has a section larger than the extruding die and considering that part of die section is occupied by fibers, solid rod speed is considerably lower than the average melt flow rate in the die. Assuming that the reinforcing fibers travel in the die at the same rate as the molten polymer, an unavoidable rate difference exists between the solid polymer and

the fibers in the feeding section. Such speed mismatch increases with increasing fiber content and induces possible friction and fiber damage problems; proper fiber bundle/die diameter ratio should therefore be selected. At the same time the polymer feeding rate should be regulated in accordance with the extrusion speed and the fiber content. From preliminary tests, it was evidenced that even few fiber ruptures in the feeding section easily lead to un-homogeneous extrusion or even die clogging and were to be avoided; thanks to their superior toughness, aramid fibers showed much better performances in this respect and were then selected for the full experimental study.

2.2 Deposition rate

A good impregnation of reinforcing fibers by a low viscosity molten polymer, is necessary to limit void content and reach optimum off-axis performances in the produced items; this suggests the use of high melt temperature. Moreover, the deposition of a continuous polymer impregnated bundle poses problems particularly when sharp corners are to be produced, due to the chance of "peeling" the deposited material before proper solidification of the polymer matrix. A selection of a relatively low extrusion rate is thus necessary to allow the full compaction soon after deposition. The feeding of the solid polymer must be properly regulated to comply with the corresponding polymer flow, in order to avoid dry fibers on one hand or polymer melt bulging on the other.

2.3 Deposition path

In the tested configuration, the reinforcing fibers are maintained continuous during the whole deposition process and no cutting system was employed. This poses limitations to the available geometries, since travel moves for the re-positioning of the depositing head without polymer extrusion should be avoided or reduced. A device for the cutting and re-attachment of the extruded material can reduce such limitations although with considerable added complexity.

The adhesion of the first deposited layer to the base is generally more problematic than in usual plain polymer deposition and can be helped with the use of a spray adhesive.

3. Experimental

3.1 Materials

Polylactic acid (PLA) was selected in this investigation in consideration of its wide employment and easy availability as filament for 3D printing. Moreover, the lower thermal shrinkage during solidification, compared to other potential materials, such as ABS, was determinant in the choice of the first tested material. Commercial PLA filament, 1.75 mm diameter was used.

As continuous reinforcing aramide fibers, Technora, HFYT-240 220dtex, were employed in the form of a yarn consisting of about 150 fibers with 12 μ m diameter each. Aramide fibers as reinforcement were selected instead of carbon of glass fibers in order to limit or prevent a possible reduction of toughness and strain at break in composite compared to PLA alone.

3.2 Extrusion head set-up

After considering a number of different approaches for the simultaneous introduction of fibers and polymer filament, a simple modification of the extruder head to accept simultaneously the reinforcement and the polymer matrix was devised. A commercial 3D printer Blue Tek Strato (provided by courtesy by Blue Tek Srl Cesano Boscone, MI, Italy) was adapted by adding continuous fibers feeding roll and using a 1mm diameter die. In fig. 1 the feeding scheme and a set-up of the system are shown.

The reinforcing fibers are fed during extrusion thanks to the pulling action of the deposited mixture. A proper control of a low deposition speed and of thin deposited layer thickness allow to obtain the necessary solidification rate, and optimal fiber impregnation during deposition. These processing parameters resulted the main factor for a proper deposition. By using aramide fibers, no significant fiber break was evidenced during the tests, so and a smooth deposition proceeds even with the use of a small diameter nozzle. Preliminary trials with glass and carbon fibers showed a higher sensitivity of such brittle fibers to premature breaks and a susceptibility to die clogging. A larger nozzle can possibly overcome such problem, yet with a consistently larger dimension of the deposited string [12].



Fig.1: Scheme (a) and set-up (b) of the adapted extrusion head of a commercial 3D printer.

It is apparent that, with such procedure, the deposition of the composite must be started by manually inserting the fibers in the die and by allowing the matrix solidification an initial point.

3.3 Composite characterization

The aramide/PLA composite obtained was characterized in terms of fiber content, compaction quality and mechanical properties. Flat bars and plates were produced by overlaying unidirectional layers with the fiber direction and dimensions required by the tests. The optimum production conditions were set after a number of deposition tests with varying parameters. All specimens were produced with a slow deposition rate, which was set at 1 mm/s and deposition thickness of 0.15 mm (0.2 mm for the first layer); the resulting width of the deposited string was 0.6-0.8 mm. Die exit temperature was set at 180 $^{\circ}$ C.

The fiber content was determined from weight and density measurements of the fibers and of produced specimens; a fiber content of 8.6 vol % (9.5 wt %) was measured.

Unidirectional specimens with 1.08 mm thickness, made of multiple layers, were observed at a tabletop scanning electron microscope SEM (Toshiba TM3000) with no surface metallization. Mechanical tests for the measure of stiffness and strength of unidirectional composites in the fiber direction (0°) and in transverse direction (90°) were carried out with an Instron 4302 dynamometer endowed with extensioneter at a crosshead rate of 2 mm/min.

Specimens for tensile tests in the fiber direction were produced with dimensions 250 mm x 15 mm x 0.95 mm (Fig.2a). End tabs were glued with cold curing adhesive for the gripping, leaving a free gage length of 150 mm. Analogously, specimen for compression tests were obtained with dimensions 60 mm x 21.3 mm x 1.5 mm and a gage length of 15 mm (Fig.2b). Specimens for tensile tests in transverse direction (90°) with dimensions 80 mm x 11 mm x 0.4 mm were cut from a large unidirectional plate (Fig2c).

Dynamic Mechanical Thermal Analysis (DMTA) was performed on unidirectional specimens with dimensions 50 mm x 7.8 mm x 1.08 mm. Temperature sweep, torsional tests at 1Hz were carried out by a Rheometrics RDA2 Analyzer.



Fig.2: Specimens for mechanical characterization: tensile (a) and compression (b) specimens in fiber direction. A large panel was manufactured for specimens to be cut and tested in transverse to fiber direction (c).

4. Results and discussion

In order to evidence limits and advantages of the devised system, a number of objects with geometries of different complexity were produced. Such tests required the setting of proper deposition parameters, thus evidencing the main issues related to practical operations. Produced items allowed also to get specimens for the mechanical and physical characterization of obtained material. Testing items produced included a cylinder, a tube segment with square section, a beam with T section, a segment with the geometry of a wing rib; moreover, rectangular bars and flat plates were also produced from which specimens for mechanical testing were cut. The issues related to the production of these objects are presented and discussed in the following.

All objects were produced with the same conditions used for the characterization specimens (deposition rate 1 mm/s, thickness 0.15 mm, die exit temperature 180 °C). Small variations of parameters were locally introduced according to object geometry.



(d)

Fig.3: Examples of the produced objects with different geometries and complexity: (a) tube with circular (30mm diameter) and (b) rectangular (80mm side) cross section, (c) beam with T section (20x14 mm) and (d) rib-shaped element (120 mm length).

The first test item is a cylinder with diameter of 30 mm and 10 mm height, made by a single layer spiral path (fig. 3a). Although the geometry is quite simple, due the small thickness and the relatively large but constant curvature some difficulty was expected. The obtained cylinder is well compacted with a fairly smooth surface, indicating that such geometry is fully compatible with the technique, provided a slow head motion is maintained.

The square section segment has sides and length of 80 mm, again made by a single layer spiral path (fig. 3b). Additional difficulty was expected in the corners, where fiber detachment due to fiber direction change and fiber "peeling" could result in the failure of operation. On the contrary, the slow deposition rate allowed a quite good overlay of the fibers with only a slight rounding of the corners.

The beam with T section has dimensions 70 mm x 20 mm x 14 mm; the thickness is 2.5 mm obtained by overlaying multiple layers (fig. 3c). The extremities of the beam, where fiber direction was reversed, were then cut and discarded, so that reinforcing fibers are unidirectional in the whole piece. The beam obtained is very good, fibers are well impregnated and regularly distributed, the section is well compacted.

A geometrical more complex example is the rib shaped element (fig. 3d) with dimensions 120 mm x 17 mm. The shape of internal stringers was designed in order to use a continuous deposition, avoiding fiber repositioning. The main difficulty introduced by the geometry is related to the sharp changes of direction, which led to slightly rounded corners and local tiny dry slots. It is expected that a more efficient cooling may further improve the obtained result. This example evidenced how a proper design of the deposition path can significantly extend the obtainable geometries.

The results of SEM inspections showed a general full compaction of the obtained composite (fig. 4), with good embedding of reinforcing fibers within the PLA matrix; only very occasional voids outside fiber bundles are observed, which, however seem not to greatly affect mechanical performances in the fiber direction.

As a matter of facts, SEM observations of tensile fractured surfaces (Fig. 4b) show that the debonding and pull-out of fibers involve rather extensive fibrillation; the pull-out lengths of fibers are estimated in the range of 0.3 to 1 mm. The images of compressive tested specimens in the failure area (Fig. 4c), show that fiber buckling is accompanied by some fiber splitting and presence of adhering material on fiber surface. These observations suggest that matrix adhesion is fair, well sufficient to significantly improve composite mechanical response.



Fig.4: SEM inspections of obtained composites: cut and polished cross-section (a), tensile (b) and compressive (c) fracture surfaces.

The mechanical tests in the fiber direction indicate that a remarkable stiffening and strengthening effect results from the addition of the reinforcing aramide. The results of tensile and compression tests are summarized in Tab.1 and in fig. 5. It can be observed that the tensile modulus of the composite is about 3 times, while tensile strength is about 6 times that of the printed PLA alone. Also maximum elongation of reinforced material is higher than that of the plain matrix; this result is opposite to what results from the addition of brittle carbon fibers [12].

Table 1 - Comparison between PLA and PLA/aramide mechanical properties.

Mechanical Properties	PLA	Composite	Variation
Tensile Modulus (fiber direction) E_1^t	3260 MPa	9340 MPa	+186%
Compressive Modulus (fiber direction) E_1^c	-	8050 MPa	
Tensile Modulus (transverse to fiber direction) E_2^t	3260 MPa	1530 MPa	-53%
Shear Modulus G_{12}	1200 MPa	1540 MPa	+28%
Tensile strength σ_U^t	34 MPa	203 MPa	+499%
Compressive strength σ_U^c	50 MPa	94 MPa	+87%
Ultimate tensile strain ε_U^t	0.028	0.033	+19%
Ultimate compressive strain ε_U^c	0.027	0.028	+1%



Fig.5: Static tensile and compressive stress-strain response of composite.

A drawback of fiber orientation and of the presence of some voids is evidenced in the transverse-tofiber direction: a modulus which is somewhat lower than that of plain PLA and a strength lower that 9 MPa were registered; also maximum elongation reduces to remarkably low values. Of course, such drawbacks can be alleviated in actual components, whenever plies with different orientations, e.g. to get cross-ply sequence, can be overlapped.

Table 2 compares mechanical properties measured in actual composites with those estimated by micromechanical models based on the mixture rule. Theoretical stiffness in the fiber direction E_1^t is directly derived from the mixture rule, while the transverse-to-fiber direction stiffness E_2^t and the shear stiffness G_{12} are obtained from the following equations [15]:

$$E_{2}^{t} = \frac{E_{f}E_{m}}{\Phi E_{m} + (1 - \Phi)E_{f}}$$
(1)

$$G_{12} = \frac{G_f G_m}{\Phi G_m + (1 - \Phi)G_f}$$
(2)

By assuming that the tensile strength of the composite in fiber direction is driven only by the reinforcing fibers, ultimate values can be easily estimated as $\sigma_c^U = \sigma_f^U \Phi$.

On the contrary, compressive failure modes involve local buckling of the fibers, matrix yielding or fiber-matrix debonding and more complex models should be adopted [16].

Values in Table 2 confirm good results obtained in fiber direction. Stiffness perfectly correlates with expected value, while the difference in terms of strength (-30.3%) can be consequence of a non-uniform distribution of stress over the fibers. Strength sensitivity to boundary conditions, edge effects, manufacturing flaws, as well as testing imperfections may have induced progressive fiber rupture, in contrast with what assumed in the estimation of material limits.

Experimental and estimated shear modulus values are also well correlated; the considerable difference of the values in the transverse to fiber direction stiffness gives relevance to reinforcement-matrix coupling and suggests that improved results should be obtained on increasing adhesion between aramide fibers and PLA.

Mechanical Property	Estimated	Experimental	Variation
Tensile Modulus (fiber direction) E_1^t	9450 MPa	9340 MPa	-1.2%
Tensile Modulus (transverse to fiber direction) E_2^t	3810 MPa	1530 MPa	-59.8%
Shear Modulus G_{12}	1550 MPa	1540 MPa	-1.3%
Tensile strength σ_U^t	290 MPa	203 MPa	-30.3%

Table 2 - Comparison between experimental and estimated mechanical properties of composite.

The thermoplastic PLA matrix has a melting temperature close to 170 °C; on the other hand, a limitation to the practical use is given by its relatively low glass transition temperature (Tg). Dynamic mechanical analysis indicates that Tg is slightly above 60 °C; above this temperature full softening of the matrix occurs and this should be considered as a maximum service limit of the material for practical uses.

5. Conclusions

Fused deposition modelling is a versatile technique to produce polymeric items with variable shapes without need of complex and costly molds. However, the polymers employed possess limited mechanical performances which can only partly improved by the addition of fillers or short fibers. In this paper, an easy adaptation of a commercial 3D printer allowed to produce continuous aramide fiber reinforced components with significant fiber content and, consequently, remarkably enhanced characteristics. The mechanical obtained performances are measured and compared to those of plain PLA. The limitation of the technique is discussed as well.

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