

## Article

# Enhancing the joining properties of biodegradable PLA from Fused Deposition Modeling by infrared laser irradiation

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**Abstract:** The development of high complexity geometry parts is one of the main goals of the additive manufacturing technology. However, the failure of printed structures and the joining of different parts to create complex assemblies represents a real challenge in the research of efficient and sustainability techniques for the permanent assembly of polymers. Laser welding processes have been used as a single step method to join metals until years ago. Nowadays, the growing trend in the use of thermoplastics for additive manufacturing has led to the need to adapt this technique to materials with a very specific nature and more sensitive to thermal effects. Also, the possibility of transmitting the laser beam through transparent polymer layers allows to focus the energy supply on internal sections of the assembled components. In this research, an infrared laser marking system was used to join two different samples of polylactic acid manufactured by fused deposited modeling technology. In order to increase the effectiveness of the bonding process, a transparent and a dark sample have been used as assembly material, focusing the laser beam on the interface area of the two parts. By means of tensile tests, dimensional measurement and the use of optical microscopy techniques, a basis was established that links the supplied energy by laser to the joining performance.

**Keywords:** Additive manufacturing; Polylactic acid (PLA); Fused deposition modelling (FDM); Laser joining; Tensile strength.

## 1. Introduction

Plastics have become attractive materials in several industrial fields through the orientation to innovative development process with lower quantity of components. This fact allows improving the assembly stages and reducing material incompatibilities. In the case of polymers for 3D printing applications, the use of new materials such as polylactic acid (PLA), with excellent thermoplastic and biodegradable properties, makes the process of great interest in a wide range of scientific areas ranging from biomechanical solutions to high impact industrial resources [1-4]. Furthermore, the possibility to improve the physicochemical and mechanical properties of PLA by the use of additives and reinforcements is one of the most investigated research lines to extend the application range of this material. Those considerations make that polylactic acid, as a biodegradable material, plays an important role to replace the petrochemical-based polymers commonly used in medical and industrial applications [5-9].

One of the main aspects of the growth in the global consume of PLA for additive manufacturing processes is based on the mechanical properties showed by this material, that can be ranging from soft and elastic elements to high strength parts. This ability to adapt the properties, can be observed in the tensile modulus range (0.35 - 3.5 GPa). In addition the relatively low melting temperature (150-162 °C) results in a highly versatile material for the development of functional parts [5].

Although a significant variety of geometries and sizes can be manufactured using PLA as based material, the complexity of some components requires the assembly of some elements through permanent or removable joining procedures. Until now, one of the most widely adopted technologies for this objective has been based on the use of chemical adhesives. However, these adhesive processes often show some characteristics that can reduce the environmental sustainability of the process [6,11,12]. As an alternative to the use of chemical adhesives, some authors suggest an innovative research line based on the joining of PLA components by the supply of focused energy over reduced areas. For this purpose, the use of laser techniques can achieve the heat transfer control for the melting of localized areas of plastic materials, facilitating the bonding process of both elements [13-17].

Laser irradiation processes have been widely used as surface modification treatments to improve the roughness of additive manufacturing (AM) parts. Through the melting of the highest asperities, the irregularities in the printing process can be reduced, resulting in a more uniform surface [18-20]. The characteristics of the laser system makes possible to concentrate high energy ranges on very small areas, increasing the accuracy of this method. In addition, the great variety of control parameter that can be used for modifying the energy supply properties to the target surface, allows to adapt the laser treatment to high specific work conditions. For this reason, laser joining technology (LJT), that includes the laser welding techniques, is particularly efficient in those applications where the chemical components of conventional adhesives can not be used (corrosive environments, biomedical components, food applications).

The nature of LJT based on light amplification implies variations in the beam transmission for different reflective materials. Under this consideration, the ability to supply the large amount of energy through transparent layers places the laser treatments in advance of other bonding technologies that needs to separate the surfaces to be joined. Although some alternatives can be performed to carry out this process, the layers of the elements subjected to the joining process usually needs to be positioning in a specific way to allow that the laser beam can pass through the upper layers. This fact implies that transparent PLA should be placed before other color layers that favors the absorption of the irradiation energy [17, 21-23]. Also, the application of thermal processes by laser avoids the need to prepare/activate the surfaces by means of chemical or mechanical procedures.

The aim of the present work was to investigate the effect of laser processing parameters on the joining properties when the PLA additive manufacturing polymer is irradiated. Nine different sets of black and colorless with variations in the energy density of pulse were tested and characterized. The joining performance were related to their maximum tensile strength after the laser irradiation process, moreover, the absence of deformation over the treated surfaces have been considered as an objective to validate the quality of this procedure. Potential application to repair damaged manufactured parts have been explored through the calculation of equivalent section dimensions for the replacement of full fill density parts.

## **2. Materials and Methods**

### *2.1. FDM printing process*

The experimental methodology to evaluate the joining properties of the laser irradiation process, considers three different probes with sizes and geometries adapted to specific evaluation conditions.

Joining probes were manufactured by Polylactic acid (PLA) filaments with  $1.75 \pm 0.03$  mm diameter from FFFWorld (Spain). A handmade modified Fused Deposition Modelling 3D printer Cube X Duo from 3D Systems (California, USA) were used. Printing parameters showed in table 1 were selected to develop upper and lower parts, under  $20 \pm 1$  °C and  $60 \pm 10\%$  relative humidity environmental conditions.

Table 1. Manufacturing parameters of printed parts

Parameter		Parameter	
Layer thickness (mm)	0.20	Filling density (%)	100
Extrusion width (mm)	0.40	Filling pattern	Linear
Extrusion temperature (K)	483.15	Feed rate (mm/s)	30
Board temperature (K)	333.15	Acceleration (mm/s <sup>2</sup> )	1000

Two different parts were designed with the dimensions showed in figure 1 to optimize the welding ability of the process, maintaining an overlapping area between upper and lower probe. In addition, to facilitate the light transmission through the upper part, focusing the beam energy on the contact area of the joining probes, transparent PLA tonality were used for the manufacturing of the upper part and black PLA for the lower part.

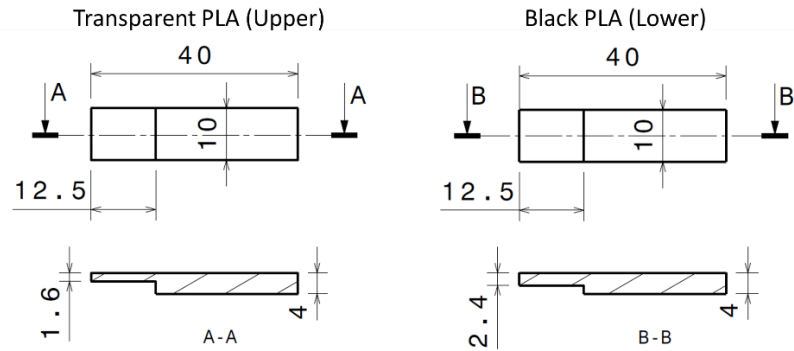


Figure 1. PLA probe design for laser irradiation joining

With the main purpose to obtain a section of a 3D printed PLA part that correspond to the axial efforts of the laser joining, a new type of probes was designed. Cylindrical samples with variation in the cross sectional area were manufactured with the same printing parameters than joining probes previously described, maintaining 100% the value of fill density.

2.2. Laser irradiation for PLA joining process

Laser joining process of the printed parts were carried out by the irradiation of a 5x5 mm area focused on the overlapping interface between the colorless and black probes, as shown in figure 2. All irradiation processes were performed using a 20 W Ytterbium-fiber infrared laser system (ROFIN-SINAR Technologies Inc., Plymouth, MI, USA). Spot diameter and pulse width were 60 μm and 100 ns respectively.

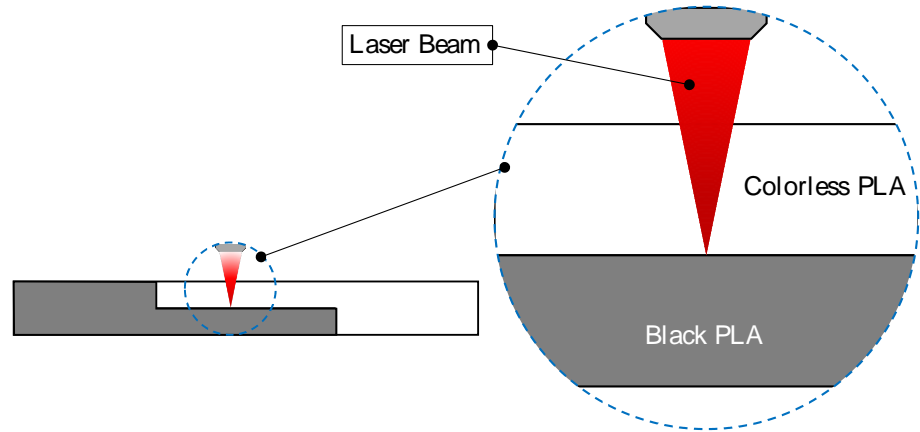


Figure 2. Laser beam focus on the interface of the colorless and black joining probes

The irradiation of the joining areas was performed at nine different pulse energy densities (Ed), using the same pulse rate and scanning speed for all the samples. Energy density was altered by selecting different laser power values, Table 2. Energy density decreases as laser power decreases, giving place to a reduction in the aggressiveness of the joining process. All the irradiation treatments were carried out through linear bidirectional layout with a 0.1 mm separation between laser tracks under open-air atmosphere.

**Table 2.** Laser irradiation parameters for joining process

Laser power (W)	Pulse rate (Hz)	Scanning speed (Vs)	Energy density of pulse (J/cm <sup>2</sup> )	Irradiated Area (mm <sup>2</sup> )
5.0	80000	50	2.21	25
6.5			2.87	
8.0			3.54	
10.0			4.42	
12.0			5.31	
13.5			5.97	
15.0			6.63	
17.5			7.74	
20.0			8.84	

On the one hand, the effects of laser irradiation process on the PLA surfaces were evaluated by optical microscopy techniques to identify specific defects of heat transfer incidence. In addition, geometrical and dimensional variations were measured using an electronic comparator set, with a 0.0001 mm resolution. Measurement of the thickness and cross-section were carried out to evaluate the incidence of the joining treatment on the resulting size of the studied parts. On the other hand, the joining strength to the failure of the laser irradiation were evaluated by standard tensile test, adapted to the PLA polymer features and the dimensions of the probes. The tensile test was performed following the UNE-EN 1465:2009 (ISO 4587:1979) standard using a Shimadzu Autograph AG-X tensile testing machine (Shimadzu Corporation, Kyoto, Japan). Specific flat grips (SCG-5KNA) for soft materials as plastics were used, with 2 mm/min test speed.

### *2.3 Corresponding size section that can be replaced by joined parts*

In order to estimate the range of the parts that can be replaced by joined sets, a new type of test probe was designed based on the size of 100% density fill of PLA cylindrical parts. These probes were developed to calculate the equivalent cross section area of a cylindrical part that can ensure the maximum tensile strength of laser irradiation joining. Taking as reference the printing parameters of the joining samples, the variation of the cross-sectional area of the cylindrical elements was taken as control parameter to evaluate the maximum tensile strength behavior as a function of the size of the printed samples. Manufacturing parameters of equivalent section cylindrical parts are shown in table 3.

**Table 3.** Manufacturing parameters of printed parts for the study of equivalent section

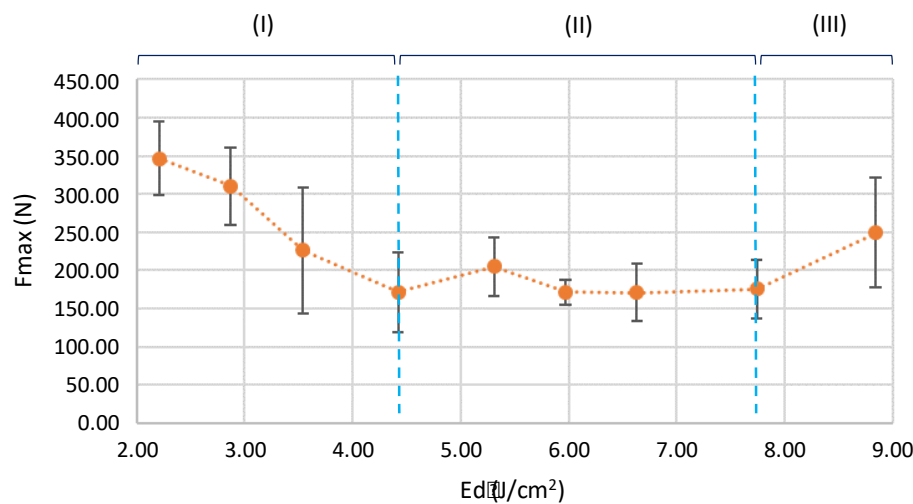
Parameter		Parameter	
Layer thickness (mm)	0.20	Filling density (%)	100
Extrusion width (mm)	0.40	Filling pattern	Linear
Extrusion temperature (K)	483.15	Feed rate (mm/s)	30
Board temperature (K)	333.15	Acceleration (mm/s <sup>2</sup> )	1000
Geometry of sample	Cylindrical	Cross section area (mm <sup>2</sup> )	7.07-9.62-12.57-15.90

### 3. Results

Although all the joined parts have failed in the overlapping area, experimental results show a significant influence of the laser radiation parameters on the properties and surface integrity of the joined samples. The use of laser techniques implies variations on the maximum tensile strength, also affecting the maximum deformation range of the probes under axial efforts. As a consequence of the high energy supplied on reduced focus area, some defects may be caused on the surface of the joining samples, decreasing the functional performance of the process.

#### 3.1. Effects of laser irradiation on tensile properties of joining parts

Energy density of pulse ( $E_d$ ) were considered as control parameter for the application of laser radiation treatments in the joining process. Increasing the  $E_d$  on the joining area results in the development of three different behavior ranges of joined parts as a function of the tensile strength. Each range is mainly characterized by a behavioral trend that provides specific characteristics to the joining process. As shown in figure 3, this trends were easily detected by tensile test of the joined samples, where the values of maximum strength ( $F_{max}$ ) and maximum deformation ( $D_{max}$ ) were affected.



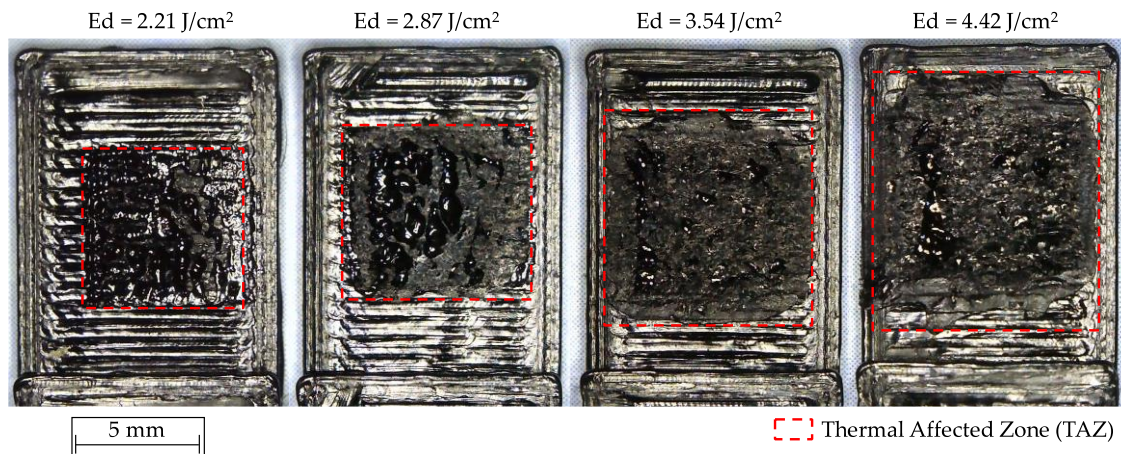
**Figure 3.** Maximum tensile strength of joined PLA as a function of energy density of pulse

The first range (I) developed under lower energy density values implies a significant decrease of the maximum tensile strength when increasing the  $E_d$ . The second set (II) observed is characterized by the uniformity of  $F_{max}$  values, maintaining a trend without significant variations in the behavior of tensile strengths. However, a slight growth can be observed in the last stage of the graph (III) for higher  $E_d$  evaluated. These variations on the maximum tensile strength behavior are highly related with the cooling process on different melted volumes of PLA under radiation treatments. The increase of supplied energy from the laser beam causes a growth of the Thermally Affected Zone (TAZ) of the contact surface of joined parts, especially in the lower sample (Black PLA), increasing the molten material volume. The higher amount of molten material generates instabilities in the cooling stage, giving rise to a lack of uniformity in the re-solidification process between contact surfaces.

#### 3.2. Effects of laser irradiation on surface integrity of joining parts

As described previously, more aggressive treatments with higher  $E_d$  values is associated with a greater incidence of the laser beam on the contact surface between the parts to be joined. As a consequence of the radiation energy supplied, a significant increase in the area of molten material considered as TAZ is caused in the lower sample, as shown in figure 4.

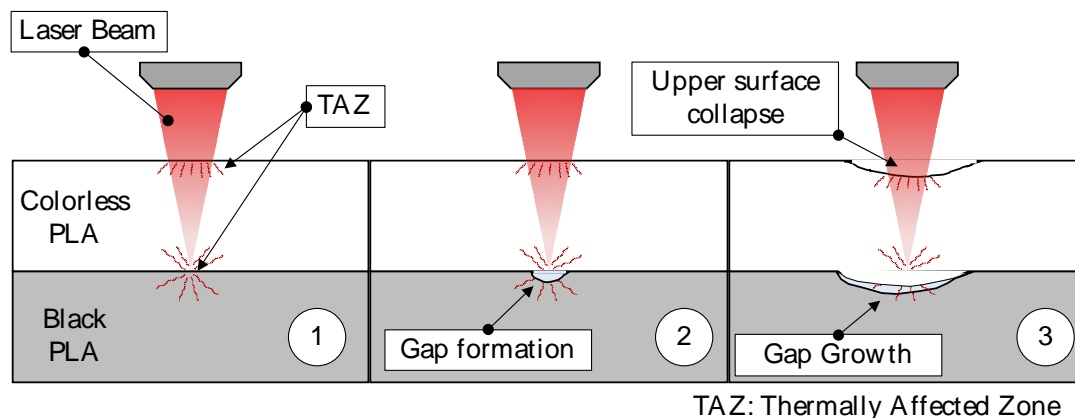




**Figure 4.** Thermally affected Zone in the lower probe (Black PLA) as a function of  $E_d$

This behavior is also associated with an increase in the depth of incidence of the beam on the lower sample, which causes the collapse of the surface and the formation of a crater. The location of the focus point of the laser beam between lower and upper joining probes also induces radiation heat transfer to each surfaces, and resulting in a deformation towards the internal layers of the black and colorless samples, as shown in stage 1 of figure 5. The increase in the supplied energy ( $E_d$ ) leads to the growth of the generated gap, reducing the joined section to the outside edge of the irradiated areas, as shown in stage 2 and 3 of the figure 5. These increases in the size and depth of craters, causes a decrease in the effectiveness of bonding zone between PLA surfaces, thus decreasing the tensile  $F_{max}$ .

For more aggressive treatments, the combined effect of heat transfer with the incidence of the beam through the colorless sample can also affect the furthest section of the probe, causing the collapse of the surface, as shown in stage 3 of figure 5. This type of defect may be considered as external defect in the joined parts, evaluated as a lack of the quality of the manufactured parts.



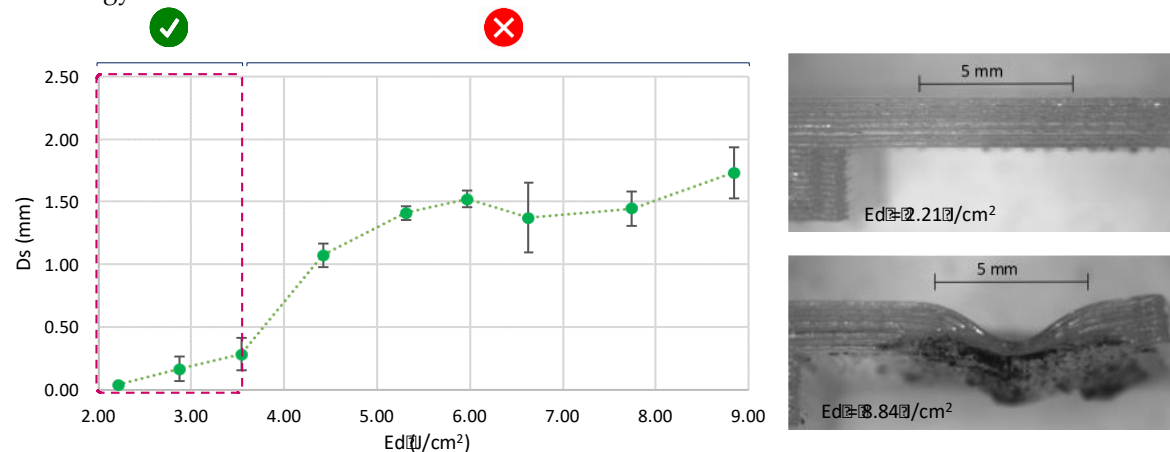
**Figure 5.** Thermal effects on the interface of the PLA samples for low, medium and high  $E_d$ .

The joining process of elements manufactured in PLA has as main objectives the assembly of complex geometries, and the repair of printed components. For this reason, the laser joining process must be carried out on reduced sections of components, and maintaining an important control of the processing parameters. This consideration leads to prevent the appearance of defects and ensure the structural integrity of the elements involved.

Upper surface collapse by irradiation effect, causes a deformation that can be considered as a quality requirement to validate the use of the technology. Under this consideration, the supplied energy, in terms of energy density of pulse of the laser treatment shows a significant influence in the

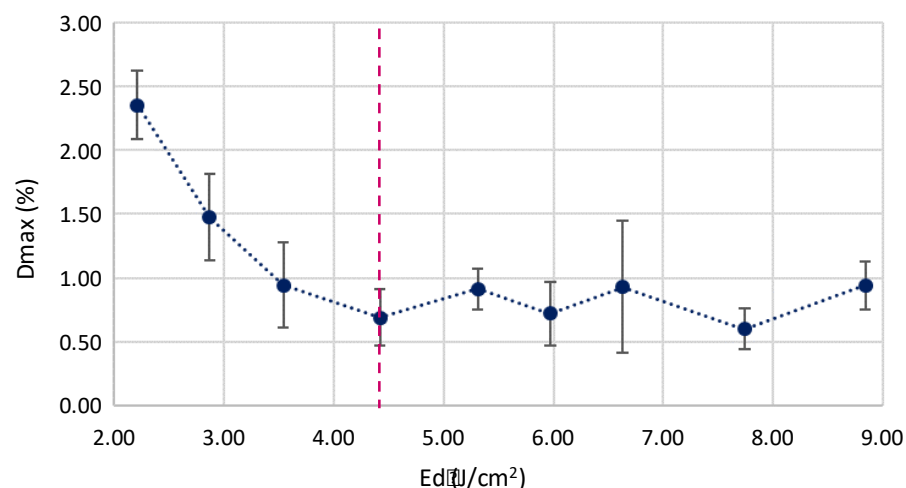
deformation of the upper surface of the joined parts. In this case, it has also been observed that increases in energy density per pulse result in greater deformation of the irradiated section, showing a critical growth from 3.54 J/cm<sup>2</sup>, as shown in figure 6.

Under this behavior, two different groups of treatments can be considered regarding to the deformation of the sample surface. On the one hand, lower energy density involves lower deformation and may be considered as not thermally affected external surfaces. On the other hand, higher energy treatments, implies an excess of the molten PLA, and as consequence, the collapse of the surface, resulting in large deformation of the external layers of the sample. As a result, may be confirmed an optimal range of pulse density energy lower than 4.00 J/cm<sup>2</sup> for the use of laser joining technology.



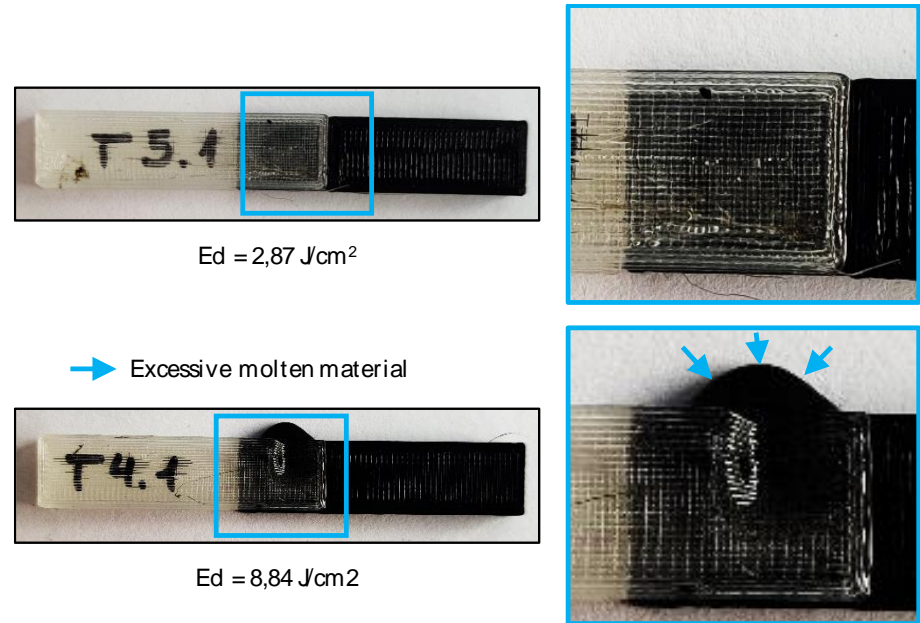
**Figure 6.** Upper surface deformation of irradiated samples for joining.

In addition to deformation phenomena on the thermally affected surfaces, the laser irradiation process may affect the printing structure of the PLA, varying mechanical properties from the initial parts. The melting and cooling process of the overlapping samples interface modify the internal printing structure of each parts, mainly based on the variation of fill density on located sections (irradiated area). One of the main consequences of the variations in the internal structure of small sections is a reduction in the tensile maximum deformation ( $D_{max}$ ). This effect is magnified when increasing the melted material volume. Based on this consideration, a significant reduction (> 220%) in the maximum tensile deformation until the failure of the joined part have been detected for lower as a function of the supplied energy ( $E_d$ ). This reduction is maintained in a uniform range for  $E_d$  values over approximately 4.42 J/cm<sup>2</sup> where critical volume of the melted material causes a similar behavior in the joined samples, as shown in figure 7.



**Figure 7.** Maximum tensile deformation of joined parts as a function of  $E_d$

Excess of melted PLA in the interlayer of the joined parts, combined with the developed gap caused by the increased process energy, causes that melting material flowing to the outside of the overlapping samples. This fact is especially evident in high energy density treatments where a significant volume of material (especially black PLA) solidifies between the joined parts, as shown in figure 8

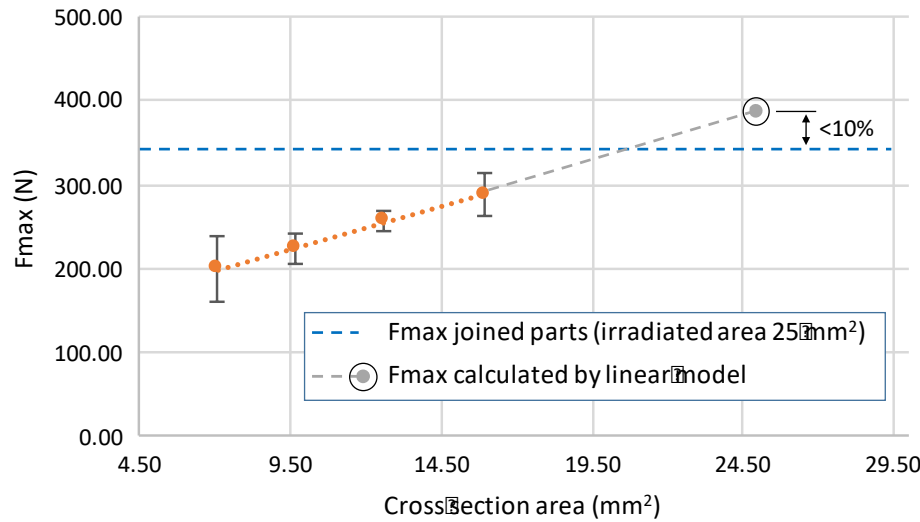


**Figure 8.** Excessive molten material for high density energy treatments

3.3. Corresponding features of joining replacing parts

One of the main purposes in the use of laser joining processes is based in the replacement and to repair PLA damaged parts. For this reason, the understanding of the equivalent sections of printed elements that can be replaced by joined parts is one of the main objectives in this research. Taking as reference the tensile strength of the 5x5 mm irradiated treatment, a study has been carried out to calculate the section of a 100% density fill printed parts that shows similar resistance than joined elements.

The evaluation of maximum tensile strength on different section printed parts has demonstrated that Fmax is highly influenced by the area of the sample, as confirmed by experimental tests. In figure 9, a linear trend has been detected, showing a proportional growth between both study parameters.



**Figure 9.** Fmax as a function of section area of equivalent printed parts



The increasing behavior of  $F_{max}$  as a function of part section ( $A$ ) is governed by the linear model of equation 1, showing a determination coefficient ( $R^2$ ) higher than 99.9% that ensure the goodness of the adjustment. Based on the linearity of the tensile strength behavior as a function of the area of printed sections, the  $F_{max}$  of equivalent irradiated area ( $5 \times 5 \text{ mm} = 25 \text{ mm}^2$ ) were calculated to validate the experimental assumptions. As a result, a variation lower than 10% were confirmed between the calculated  $F_{max}$  of the equivalent sections, of full density fill printed parts, and the  $F_{max}$  value of the joined parts that fits with the joining conditions of  $E_d=2.21 \text{ J/cm}^2$ .

$$F_{max} [\text{N}] = 10.124 \cdot A [\text{mm}^2] + 127.160 \quad (1)$$

Proposed model, and its validation by experimental procedures, allows to detect the optimum values of the laser irradiation parameters to ensure the highest resistance and minimum deformation of the joined parts. Under these processing conditions, all the tested samples show similar tensile strength to continuous full density fill printed parts, obtaining a first approximation to the calculation of replacement sections for damaged parts by laser joining.

## 5. Conclusions

The effect of laser processing parameter on the thermal joining behavior of polylactic acid from fused deposition modelling technology have been studied. Three different behavioral ranges of joined parts have been confirmed, in terms of maximum tensile strength ( $F_{max}$ ), as a function of the supplied energy density of pulse ( $E_d$ ), reaching a maximum value of 346.54 N for 2.21  $\text{J/cm}^2$  of  $E_d$ . Although a stabilization of the values of  $F_{max}$  have been detected from 4.42  $\text{J/cm}^2$  energy density, a significant increase in the thermally affected area as a consequence of the incidence of the beam for higher  $E_d$  values. Increases of the thermally affected zone produces an internal gap formation between assembled parts that may cause the collapse of the upper surface, mainly due to the excess of melting material. In addition, cooling processes of higher volumes of melting materials may reduce the tensile maximum deformation by up to 220%. Under this consideration optimal conditions for joining process by infrared laser on transparent and black PLA parts have been obtained for the lower energy density tested, where higher  $F_{max}$  and lower deformation of external surfaces were observed. Based on the linear behavior of the cross sectional area of PLA manufactured parts under tensile efforts, in terms of  $F_{max}$ , may be confirmed that laser joined sections of 25  $\text{mm}^2$  have showed similar tensile resistance (<10%) than 25  $\text{mm}^2$  cross sectional parts (100% fill density).

**Author Contributions:** Conceptualization, JMVM and DPV.; methodology JMVM and DPV; validation, JMVM, MBP and DPV; investigation, JMVM, JSG and MBP; writing—original draft preparation, JMVM, writing—review and editing, JMVM, MBP, JSG and DPV. All authors have read and agreed to the published version of the manuscript.

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