**From disposal to sustainable development: technological potential of poly(lactic acid) (PLA) blends with 3D filament waste**

**Do descarte ao desenvolvimento sustentável: potencial tecnológico de blendas de poli(ácido lático) (PLA) com resíduos de filamentos 3D**

**De la eliminación al desarrollo sostenible: potencial tecnológico de las mezclas de poli(ácido láctico) PLA con residuos de filamentos 3D**

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**Abstract**

Additive manufacturing is growing rapidly in the automotive, medical, and aerospace industries as an option for the manufacturing of products. However, there is a continuous growth in the amount of waste generated by 3D filaments, thus, the reuse practice becomes important, since it brings environmental and economic gains. The present research evaluated the mechanical, thermal, thermomechanical and rheological properties of PLA/PLAr blends containing post-consumption 3D filament. The blends were prepared in a co-rotational twin screw extruder and, subsequently, the extruded granules were injection molded. As the PLAr content in the blends (PLA/PLAr) increased, there was a reduction in viscosity, indicating an improvement in manufacturability. The PLA/PLAr blend (75/25 % wt.) increased the degree of crystallinity compared to neat PLA, indicating that PLAr acted as a nucleating agent. As a consequence, the PLA/PLAr blend (75/25 % wt.) showed performance comparable to neat PLA in thermal stability, elastic modulus, tensile strength, Shore D hardness, impact strength, heat deflection temperature (HDT) and Vicat softening temperature. The reuse of post-consumption 3D filament PLA is feasible for the development of materials with good properties. In addition, value is added to the post-consumption material and there is a contribution to sustainable development.

**Keywords:** 3D filaments; Waste; Reuse; Poly(lactic acid) - PLA; Polymer blends.

**Resumo**

A manufatura aditiva está crescendo de forma acelerada para a fabricação de peças nas indústrias automotiva, médica e aeroespacial. Porém, há um crescente aumento no teor de resíduos gerados pelos filamentos 3D e, portanto, a prática do reaproveitamento torna-se importante, uma vez traz ganhos ambientais e econômicos. A presente pesquisa avaliou as propriedades mecânicas, térmicas, termomecânicas e reológicas de blendas de PLA/PLAr pós-consumo de filamentos 3D. As blendas foram preparadas em uma extrusora de rosca dupla corrotacional e, posteriormente, os grânulos extrudados foram moldados por injeção. À medida que aumentou o teor de PLAr nas blendas (PLA/PLAr) houve uma redução na viscosidade, indicando um melhoramento na processabilidade. A blenda PLA/PLAr (75/25 % em massa) aumentou o grau de cristalinidade em comparação ao PLA virgem, indicando que o PLAr atuou como agente nucleante. Como consequência, a blenda PLA/PLAr (75/25 % em massa) apresentou desempenho equiparável ao PLA puro na estabilidade térmica, módulo elástico, resistência à tração, dureza Shore D, resistência ao impacto, temperatura de deflexão térmica (HDT) e temperatura de amolecimento Vicat. O reaproveitamento do PLA pós-

consumo de filamentos 3D é viável para o desenvolvimento de materiais com boas propriedades. Além disso, agrega-se valor ao material pós-consumo e contribuindo para o desenvolvimento sustentável.

**Palavras-chave:** Filamentos 3D; Resíduos; Reaproveitamento; Poli(ácido lático) - PLA; Blendas poliméricas.

**Resumen**

La fabricación aditiva está creciendo rápidamente para la fabricación de piezas en las industrias automotriz, médica y aeroespacial. Sin embargo, existe un aumento creciente en el contenido de residuos generados por los filamentos 3D y, por tanto, la práctica de la reutilización se vuelve importante, ya que aporta beneficios medioambientales y económicos. La presente investigación evaluó las propiedades mecánicas, térmicas, termomecánicas y reológicas de mezclas de PLA/PLAr posconsumo de filamentos 3D. Las mezclas se prepararon en una extrusora de doble husillo rotacional y, posteriormente, los gránulos extruidos se moldearon por inyección. A medida que aumentó el contenido de PLAr en las mezclas (PLA/PLAr), hubo una reducción de la viscosidad, lo que indica una mejora en la procesabilidad. La mezcla de PLA/PLAr (75/25 % en masa) aumentó el grado de cristalinidad en comparación con el PLA virgen, lo que indica que el PLAr actuó como agente nucleante. Como consecuencia, la mezcla PLA/PLAr (75/25 % en masa) mostró un rendimiento comparable al PLA puro en estabilidad térmica, módulo elástico, resistencia a la tracción, dureza Shore D, resistencia al impacto, temperatura de deflexión térmica (HDT) y temperatura de ablandamiento Vicat. La reutilización de PLA de filamentos 3D postconsumo es factible para el desarrollo de materiales con buenas propiedades. Además, se agrega valor al material de posconsumo y se contribuye al desarrollo sostenible.

**Palabras clave:** Filamentos 3D; Residuos; Reutilización; Poli(ácido láctico) - PLA; Mezclas de polímeros.

## 1. Introdução

Polymeric materials make up a large part of the waste generated by humanity, and are often improperly discarded in the environment, causing serious environmental and social impacts (Carmo et al., 2020; Okan et al., 2019; Luna et al., 2020a). These problems are aggravated by the fact that commodity polymers (polyethylene, polypropylene, poly (vinyl chloride) and polystyrene) are produced from oil, generating high durability and resistance to

biodegradation (Valerio et al., 2020; Camargo et al., 2020; Luna et al., 2016). As an alternative to reduce the environmental impact of commodity polymers, a new class of materials has emerged, biopolymers, which include "biodegradable or ecologically correct materials" (Galindo et al., 2020; Shetty and Shetty, 2019; Morais et al., 2019; Siqueira et al., 2019a).

Poly(lactic acid) - PLA is a biodegradable polymer obtained from renewable sources, commercialized and produced worldwide on a large scale, with an estimated annual increase of 10% to 15%, while the world market for plastics has grown from 3.5% to 4% per year (Oliveira et al., 2020). PLA has awakened great academic and industrial interest since it has mechanical properties close to other commercial polymers (Lima et al., 2016).

PLA is an aliphatic polyester, thermoplastic, with polar chemical structure, with the possibility of being semicrystalline or amorphous depending on the processing conditions (Wang et al., 2018; Chaiwutthinan et al., 2019). From the mechanical behavior point of view, PLA has good tensile strength and elastic modulus, but has low impact strength (Brito et al., 2012). In addition, the characteristics of transparency, bio absorbability, biocompatibility, and manufacturability, suggest potential for several applications, from packaging to more specialized applications such as biomedical (Araújo et al., 2015). Currently, PLA is widely used in the area of additive manufacturing (3D printing), due to its ease of use and the good results for the manufactured parts (Wang et al., 2018; Balani et al., 2019). As a consequence, the demand for PLA has increased in the market for the manufacture of 3D printed parts, generating waste with the potential for reintroduction in the production chain. The reuse of post-consumption PLA becomes of great importance, since the simple disposal in the environment or in dumps leads to the waste of raw material with good properties and high added value (Andrade et al., 2016; Piemonte et al., 2013). The practice of reusing polymers contributes directly to sustainability, as it establishes a more harmonious relationship between the industry, the consumer of plastic materials and the environment (Luna et al., 2020b; Zander et al., 2019). However, there is a need to investigate the effect of reprocessing on the mechanical properties of PLA, so the content of recycled material in the composition with neat PLA can be controlled in order to obtain acceptable properties and a quality product (Babagowda et al. 2018).

Hong et al. (2020) investigated the effect of mechanical recycling on PLA filament for 3D printing, aiming to evaluate mechanical and thermal properties. The glass transition temperature ($T_g$), the cold crystallization temperature and the degree of crystallinity of the reprocessed PLA is less than what is found in virgin PLA, contributing to a decrease in tensile

and flexural strengths. The authors indicated the need to control the reprocessing cycle in order avoid severe deterioration of the mechanical properties.

The recycling and reuse of most commodity polymers have been extensively studied over the years (Satya et al., 2020; Luna et al., 2019a; Luna et al., 2015; Luna et al., 2014; Araújo and Morales, 2018; Ferreira et al., 2019a; Aishah et al., 2020), indicating the feasibility of developing blends between virgin and recycled polymers, in controlled quantities. Meanwhile, blends of poly (lactic acid) and PLA waste from 3D filaments have practically not been investigated in polymer literature, thus leaving a gap to be investigated on the reuse of PLA from additive manufacturing. In view of the importance of PLA for the development of ecological materials and for the 3D manufacturing industry, research on the potential of this post-consumption polymer becomes justified, contributing to the reuse, increase in value and development of the sustainable cycle.

The objective of this work was to analyze the mechanical, thermal, thermomechanical, and rheological properties of PLA blends with post-consumption PLAr of 3D filaments (25%; 50% and 75% wt.).

## 2. Materials and Methods

**Materials**

Poly(lactic acid) (PLA) with melt flow index (MFI) = 6 g/10 min (210 ºC/2.16 kg) and density = 1.24 g/cm³, in the form of pellets and manufactured by Nature Works, distributed by 3D Lab (Brazil).

The poly(lactic acid) waste (called PLAr, where "r" refers to recycled) from 3D printer filaments, used to make parts and plates. These filaments contain pigments to modify the color and are sold by 3D Lab.

**Methods**

*Recycled PLA preparation*

The recycled PLA waste (PLAr) was crushed in a knife mill grinder, resulting in flakes. Subsequently, these flakes were washed with water, in order to remove contaminants and then, dried in a vacuum oven for 24 hours, at a temperature of 60 °C.

*Extrusion processing*

The PLA/PLAr blends were dry-mixed and then processed in a modular co-rotational twin screw extruder, model Zweiwelliger SchneckenKneter ZSK (D = 18 mm and L/D = 40), from Coperion Werner-Pfleiderer (Stuttgart-Feuerbach, Germany). The temperature profile used in the extruder was 180 °C - 180 °C - 180 °C - 180 °C - 180 °C - 180 °C - 190 °C, screw rotation speed of 250 rpm and controlled feed rate of 3 kg/h, with screw profile containing elements of distributive and dispersive mixture. The granulated material was dried in a vacuum oven for 24 hours, at a temperature of 60 °C. The neat PLA was processed and dried in the same conditions as the blends (PLA/PLAr). Table 1 shows the compositions found in the blends, based on mass proportions (%).

**Table 1**. Blend composition by mass proportion (%).

| Sample | PLA (% wt.) | PLAr (% wt.) |
|---|---|---|
| PLA | 100 | - |
| PLA/PLAr | 75 | 25 |
| PLA/PLAr | 50 | 50 |
| PLA/PLAr | 25 | 75 |
| PLAr | - | 100 |

Source: Authors.

*Injection molding*

The blends and neat PLA were injection molded in an Arburg injection molding machine (ARBURG Technology Center, Kaiserstr, Radevormwald, Germany), Model Allrounder 207C Golden Edition, to obtain impact (12.8 x 3.2 x 64 mm), traction (12.9 x 3.2 x 165 mm) and HDT (12.8 x 3.2 x 125 mm) specimens, according to ASTM D256, ASTM D638 and ASTM D648 standards, respectively. The molding conditions are shown in Table 2.

**Table 2**. Specimen injection molding conditions.

| Parameters | Condition |
|---|---|
| Injection pressure (bar) | 1000 |
| Temperature profile (°C) | 185, 185, 185, 185 e 185 |
| Mold temperature (°C) | 20 |
| Mold cooling time (s) | 30 |
| Back pressure (bar) | 500 |

Source: Authors.

Figure 1 shows a schematic representation of the processing of neat PLA and blends, by extrusion and injection.

**Figure 1**. Schematic representation of extrusion and injection processing.
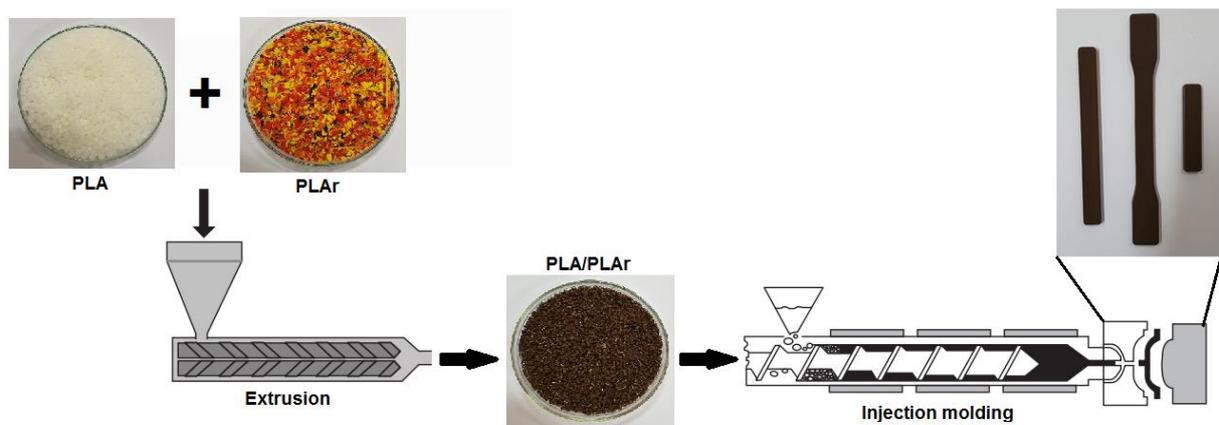


Source: Adapted from Brito et al. (2012).

*Material Characterization*

The melt flow index (MFI) measurements were performed on a plastometer, model DSM MI-3 (Franca, Brazil), according to the ASTM D-1238 standard, with a load of 2.16 kg and a temperature of 210 °C. The results were analyzed as an average of ten specimens.

The Izod impact strength test was performed on notched specimens, according to the ASTM D256 standard, in a Ceast model Resil 5.5 J device (Turin, Italy), operating with a 2.75 J hammer, at room temperature (~ 23 °C). The results were analyzed as an average of six specimens.

The tensile test was performed on injected specimens, according to the ASTM D638 standard, using a universal testing machine of the EMIC DL 2000 brand (São José dos Pinhais, Brazil), with a speed of 5 mm/min and a load cell of 20 kN, at room temperature (~ 23 °C). The results were analyzed as an average of six specimens.

The penetration resistance was determined according to the ASTM D2240 standard, on a Shore "D" durometer (São Paulo, Brazil), with a load of 50 N. The results were analyzed as an average of seven penetrations.

The heat deflection temperature (HDT) was obtained according to the ASTM D648 standard, in a Ceast equipment, model HDT 6 VICAT (Turin, Italy), with a tension of 1.82 MPa and a heating rate of 120 °C/h. The temperature was determined after the sample was deflected 0.25 mm. The results were analyzed as an average of three specimens.

The Vicat softening temperature was performed according to the ASTM D1525 standard, in a Ceast equipment, model HDT 6 VICAT (Turin, Italy), using a heating rate of 120 °C/h and a load of 50 N. The temperature was determined after the needle penetrated 1 mm in the specimens. The results were analyzed as an average of three specimens.

Differential scanning calorimetry (DSC) analysis was performed on a DSC-Q20 from TA Instruments (New Castle, EUA). The test was carried out starting from 30 to 200 °C, with a heating rate of 10 °C/min, 3 min isotherm ramp, gas flow of 50 mL/min, nitrogen atmosphere and using 6 mg of specimen.

The thermogravimetric (TG) analysis were obtained in a simultaneous TG/DSC equipment from TA Instruments, model SDT Q600 (New Castle, EUA), employing about 6 mg of the specimen, with a heating rate of 10 °C/min and gas flow of 100 mL/min, starting from room temperature 30 to 500 °C, in a nitrogen atmosphere.

The contact angle analysis was performed using the sessile drop method, with a portable contact angle, Phoenix-i model from Suface Eletro Optics – SEO (Gyeonggi-do, South Korea). The drop was deposited on an impact test specimen using a micrometric dispenser, the image being captured and analyzed by the equipment's own software.

**3. Resultados e Discussão**

*Melt Flow Index (MFI)*

The melt flow index (MFI) has been widely used in the industry to characterize flow properties of polymers, due to the simplicity and quickness of the test (Rocha et al., 1994). To estimate the viscosity of neat PLA, PLAr and blends (PLA/PLAr), the melt flow index technique was utilized, which is inversely proportional to viscosity, which in turn is directly related to molecular weight (Luna et al., 2019b). Thus, the higher the melt flow index, the lower the viscosity.

In Figure 2, the results of the melt flow index of neat PLA, PLAr and blends (PLA/PLAr) are presented, according to the content of recycled material.

**Figure 2**. Melt flow index of neat PLA, PLAr and blends (PLA/PLAr).
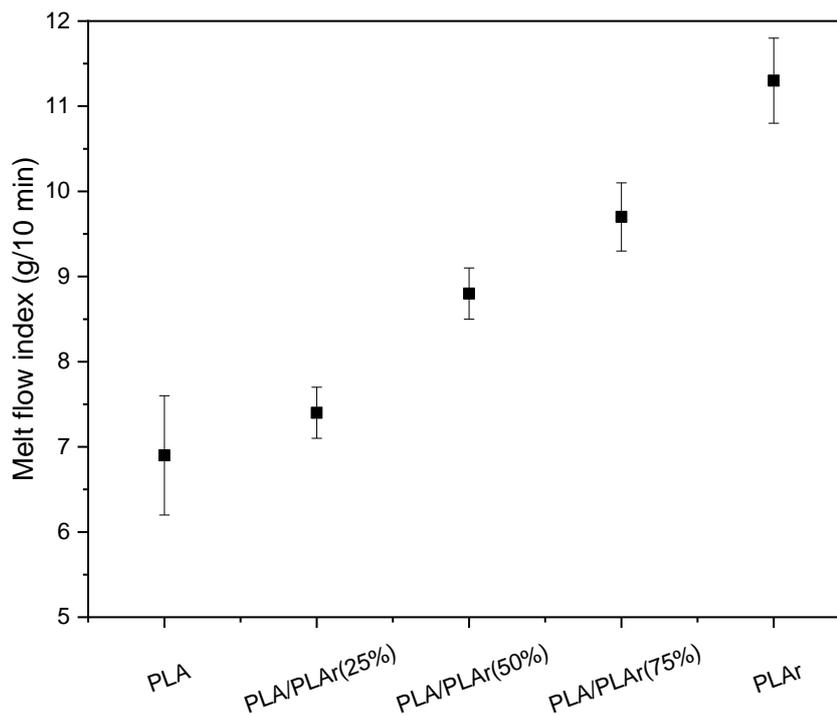


Source: Authors.

PLA had the lowest melt flow index (6.9 g/10 min) and PLAr the highest fluidity rate (11.3 g/10 min), indicating greater and lesser molar mass, respectively. The PLAr has already been subjected to high temperature processing during the 3D printing and, as a consequence, has undergone chain scission, causing a reduction in molar mass, increasing fluidity, and

reducing viscosity (Aguero et al., 2019). The binary blends (PLA/PLAr) showed intermediate melt flow index values to those of the respective pure polymers (PLA and PLAr), as expected by the additivity. As the PLAr content was increased to the neat resin, the flow rate increased, being more pronounced when using 75% PLAr. The incorporation of recycled material (PLAr), with smaller molar mass, to neat PLA favored a plasticizer effect, contributing to reduce viscosity.

*Differential Scanning Calorimetry (DSC)*

Table 3 provides the fusion, crystallization, and glass transition parameters of neat PLA, PLAr and blends (PLA/PLAr), respectively. The DSC curves obtained during the second heating cycle can be seen in Figure 3.

**Table 3**. Fusion and crystallization parameters of PLA, PLAr and blends (PLA/PLAr), respectively. *$T_g$ = glass transition temperature; $T_m$ = peak melting temperature; $T_c$ = Crystallization temperature; $T_{cf}$ = Cold crystallization temperature; $\Delta H_f$ = Enthalpy of fusion; $X_c$ = Degree of crystallinity, $X_c = \Delta H_f / \Delta H_0$, where: $\Delta H_0$ = Enthalpy of fusion of PLA with 100% crystallinity, 93.7 J/g (Silva et al., 2014).

| Specimen* | $T_g$ (°C) | $T_m$ (°C) | $\Delta H_m$ (J/g) | $T_{cf}$ (°C) | $T_c$ (°C) | $X_c$ (%) |
|---|---|---|---|---|---|---|
| PLA | 60.7 | 177.6 | 53.3 | 92.8 | 95.0 | 56.8 |
| PLA/PLAr (25%) | 59.8 | 178.3 | 60.2 | - | 98.4 | 64.2 |
| PLA/PLAr (50%) | 59.4 | 176.1 | 48.0 | - | 100.1 | 51.2 |
| PLA/PLAr (75%) | 59.4 | 174.4 | 44.9 | - | 100.5 | 47.9 |
| PLAr | 59.1 | 172.9 | 41.1 | - | 99.6 | 43.8 |

Source: Authors.

**Figure 3**. DSC curves for neat PLA, PLAr and blends (PLA/PLAr): (a) melting temperature and (b) crystallization temperature.
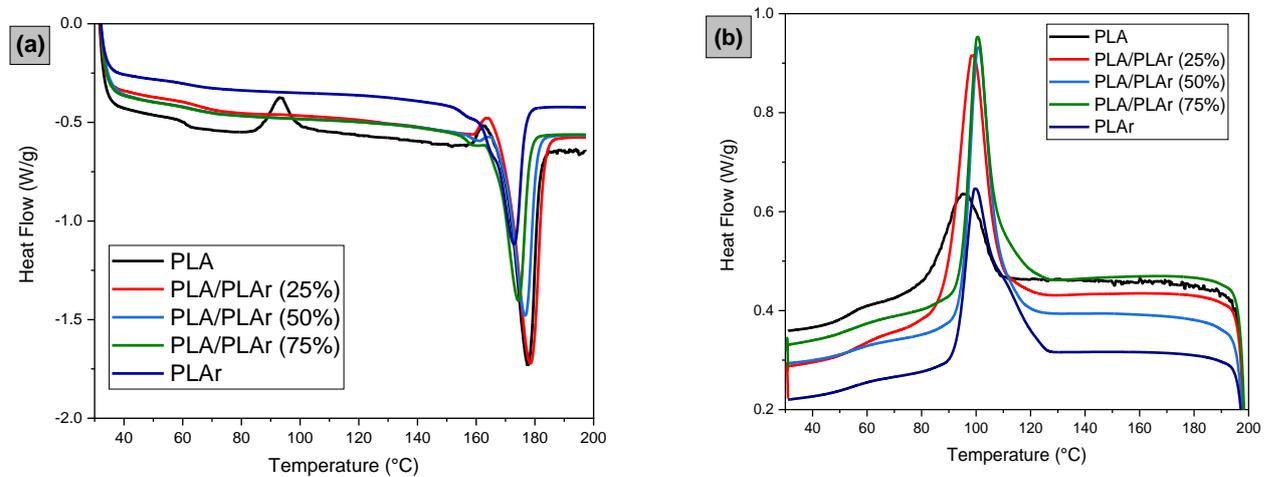


Source: Authors.

The neat PLA had a glass transition temperature ($T_g$) of approximately 60.7 °C, while the PLAr around 59.1 °C, values close to those reported in the literature (Boubekeur et al., 2020; Santana et al., 2018). The PLA/PLAr blends showed a slight shift in $T_g$ for lower temperatures, when compared to neat PLA. This indicates that the addition of recycled PLA contributed to the corresponding reduction in $T_g$, suggesting a plasticizer effect. As the PLAr presented a lower molar mass, as verified in the melt flow index, there is a reduction in the energy required for the molecular movement (Maiza et al., 2015), generating a decrease in $T_g$.

In Figure 3, an exothermic peak is observed during the heating of the neat PLA, due to cold crystallization process, a phenomenon observed in the literature for the PLA (Yu et al., 2008). This phenomenon occurs above $T_g$, when PLA molecules acquire sufficient mobility to rearrange themselves in a crystalline structure (Di Lorenzo et al., 2013). Cold crystallization renders materials with better mechanical and barrier properties, but compromises transparency (Wellen and Rabello, 2007). PLAr and blends did not show the cold crystallization peak, indicating that the addition of PLAr inhibited the process. This indicates that the presence of PLAr with lesser molar mass makes the molecular structure less regular and, therefore, less prone to cold crystallization during heating (Wellen and Rabello, 2008).

The neat PLA presented an endothermic event around 177.6 °C, referring to the crystalline fusion temperature ($T_m$) of the α phase (Mohapatra et al., 2012; Fukushima et al., 2009). The melting temperature of the PLAr was around 172.9 °C, lower when compared to

the PLA's. As the reduction in the melting temperature is often associated with the crystal's degree of perfection (Nascimento et al., 2013), it is suggested that the lower molar mass of the PLAr has nucleated smaller and less thick crystals, reducing its degree of perfection. Apparently, the 25% PLAr content is a critical concentration to induce the nucleation of more perfect crystals, since the melting temperature shifted to 178.3 °C, when compared to PLA. However, when increasing the content to 50% and 75% of PLAr in the blends (PLA/PLAr), there is a continuous tendency to reduce the melting temperature. In this case, clearly, the recycled PLA contributed to reduce the perfection of the crystals in the virgin resin (PLA) and, consequently, needing lower temperatures to melt the crystals. Pillin et al. (2008) observed a reduction in the melting temperature of the PLA, indicating that each processing cycle induces degradation in the PLA and produces chain scission, with reduced viscosity and generation of less perfect crystals.

In Figure 3, it can be seen that before the main endothermic peak for neat PLA and PLA/PLAr blends (25%; 50% and 75%), there is a small deflection due to the growth of a small crystal population (Di Lorenzo et al., 2011). Regarding the enthalpy of fusion $\Delta H_f$, the PLA required 53.3 J/g to melt the crystals, while less energy was required by the PLAr (41.1 J/g). The addition of 50% and 75% of PLAr in the blends (PLA/PLAr) progressively reduced the enthalpy of fusion, compared to neat PLA. In this case, blends with 50% and 75% recycled PLA require less energy to melt the crystals. However, when 25% of recycled PLA is added to neat PLA, there is a greater need for energy ($\Delta H_f$) to melt the crystals, compared to other blends. Consequently, the 25% PLA/PLAr blend impacted a greater intensity and width of the fusion peak, indicating that the global crystallinity increased, in relation to the neat PLA.

The PLA had a crystallization temperature ($T_c$) of around 95 °C, while the PLAr had a higher temperature of around 99.6 °C. The PLA/PLAr blends showed a higher crystallization temperature compared to PLA. Such behavior indicates that the incorporation of PLAr favored the initiation of crystallization at higher temperatures than neat PLA. Apparently, as PLAr is a material with added pigments, there is tendency for the formation of crystallization nuclei, reducing the critical free energy for the consolidation of the nucleus and accelerating the global crystallization process (Rabello, 2000).

The degree of crystallinity of the PLA was in the order of 56.8%, a value higher than that found for the PLAr (43.8%). The addition of PLAr in high proportions (50% and 75%) to virgin PLA had an impact on reducing the degree of crystallinity, when compared to PLA. This indicates that the formation of neat PLA crystals was hindered by the presence of

recycled PLA in high proportions, reinforcing the reduction in the melting temperature. However, when using 25% of PLAr in the composition of the PLA/PLAr blend, a different behavior occurred, with a degree of crystallinity in the order of 64.2%, higher than the neat PLA. Apparently, the 25% PLAr content is acting as a nucleating agent, generating an increase in the degree of crystallinity ($X_c$) and in the crystallization temperature ($T_c$), indicating that the PLA/PLAr blend (75/25%) had a synergistic effect.

### *Thermogravimetry (TG)*

Figure 4 (a, b) illustrates the TG and DTG curves of neat PLA, PLAr and blends (PLA/PLAr), according to the content of recycled material, respectively.

**Figure 4**. Thermal behavior of neat PLA, PLAr and blends (PLA/PLAr): (a) thermogravimetric curves; (b) first derivative (DTG).
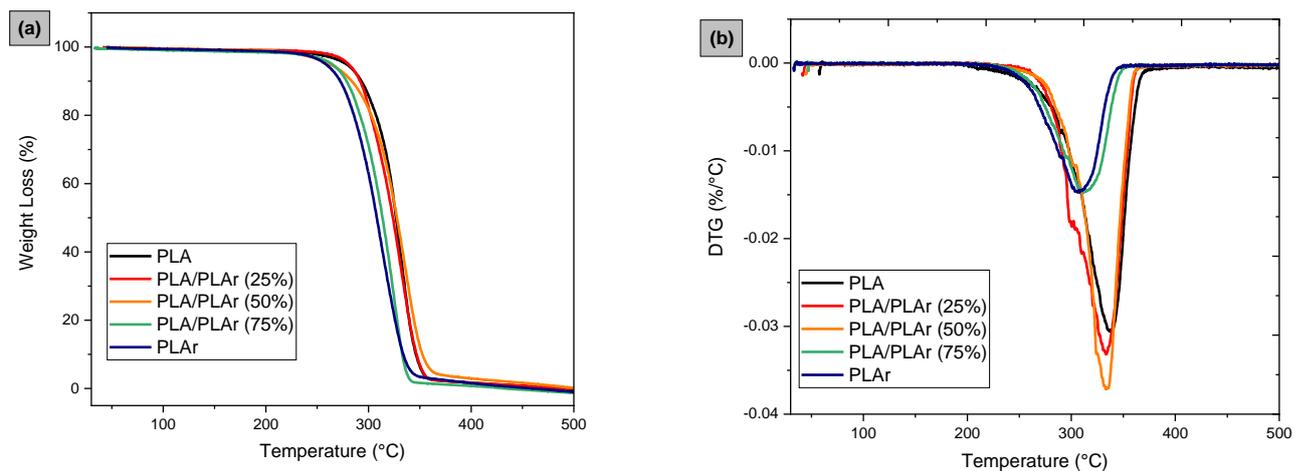


Source: Authors.

In Figure 4(a), the PLA presented a single thermal decomposition step in the range of 300 to 400 °C, due to the process of chain degradation (Silva et al., 2020). It is possible to observe for PLAr a single decomposition step initiated within the range of 250 to 352 °C, indicating that it presented less thermal stability, when compared to PLA. As the PLAr showed lower molar mass and degree of crystallinity, thermal stability decreased (Manaf et al., 2018; Becena, 2005). The PLA/PLAr blend (25%) hardly changed the thermal decomposition behavior, when compared to neat PLA. This indicates that when using 25% of PLAr in the formulation, a synergistic effect occurred, with interactions that did not cause a

deteriorating effect on thermal stability. With the increase in the PLAr content to 50% and 75% in the PLA/PLAr blends, there is a shift in the curves to lower temperatures, implying a reduction in thermal stability. However, for the blend with 75% PLAr the effect of the instability on the thermal behavior is more pronounced, with a tendency to approximate the PLAr curve.

In Figure 4(b) and Table 4, the peaks of the DTG curves are observed, corresponding to the temperature at which the maximum rate of mass loss occurs (Luna et al., 2020c). PLA and PLA/PLAr blends (25 and 50%) showed similar degradation temperatures since the curves are practically overlapping. At the same time, it can be noted that there is no noticeable change in the mechanism of thermal degradation of the PLA and the PLA/PLAr blend (75%), as the curves of the first derivative are practically identical in shape and intensity. In addition, there is a reduction in the peak of the DTG for lower temperatures, suggesting a decrease in the thermal stability of the PLAr and the PLA/PLAr blend (75%), when compared to the other materials. Such behavior confirms that the PLAr needs less energy to cause the fusion process, corroborating with the DSC.

Table 4 shows the temperature at which 10% loss of mass occurs ($T_{0.1}$), considered as the initial temperature of the thermal degradation process (Ribeiro et al., 2012). PLA lost mass at a higher temperature when compared to PLAr, indicating that the former is more thermally stable. As PLAr is contains organic pigments, the 10% loss of mass started at a lower temperature (272.3 °C). The PLA/PLAr blend (25%) had a 10% loss in mass at a temperature similar to PLA, which indicates similar thermal stability. As the PLAr content increased (50 and 75%) in the blends, there was a drop in the 10% mass loss temperature, that is, the recycled material imposes a reduction in thermal stability, when compared to PLA. The results of $T_{0.1}$ show that the thermal decomposition processes of the blends (PLA/PLAr) stay intermediate to the degradation processes of pure polymers (PLA and PLAr). In general, for a lower PLAr content (25%) there was a synergistic effect without compromising thermal stability, as seen in the DSC analysis.

**Table 4**. Results for 10% loss of mass ($T_{0.1}$) and the maximum temperature of mass loss ($T_{dm}$).

| Specimen | $T_{0.1}$ (°C) | $T_{dm}$ (°C) |
|---|---|---|
| PLA | 292.8 | 338.7 |
| PLA/PLAr (25%) | 291.6 | 334.6 |
| PLA/PLAr (50%) | 283.6 | 334.2 |
| PLA/PLAr (75%) | 278.5 | 312.3 |
| PLAr | 272.3 | 306.9 |

Source: Authors.

### *Izod Impact Strength*

Figure 5 shows the Izod impact strength of neat PLA, PLAr and blends (PLA/PLAr), according to the content of recycled material, respectively. The PLA presented an impact strength in the order of 29.1 J/m, typical value of polymers with fragile behavior (Brito et al., 2012). The performance in the impact strength of PLAr was 27.3 J/m, corresponding to a slight reduction of 6.2%, when compared to PLA. As the PLAr had a lower molar mass, a reduction in impact strength was expected, but it was not a drastic decrease in this property. This is due to the fact that PLAr has undergone a primary mechanical recycling, contributing to a lack of severe degradation (Spinacé and Paoli, 2005) and generating properties similar to neat PLA. For the PLA/PLAr blends, it appears that the PLAr concentration's influence is insignificant to change the impact strength. The concentrations used (25%; 50% and 75%) provide similar behaviors, since they are within the margin of experimental error.

**Figure 5**. Izod impact strength of PLA, PLAr and blends (PLA/PLAr).
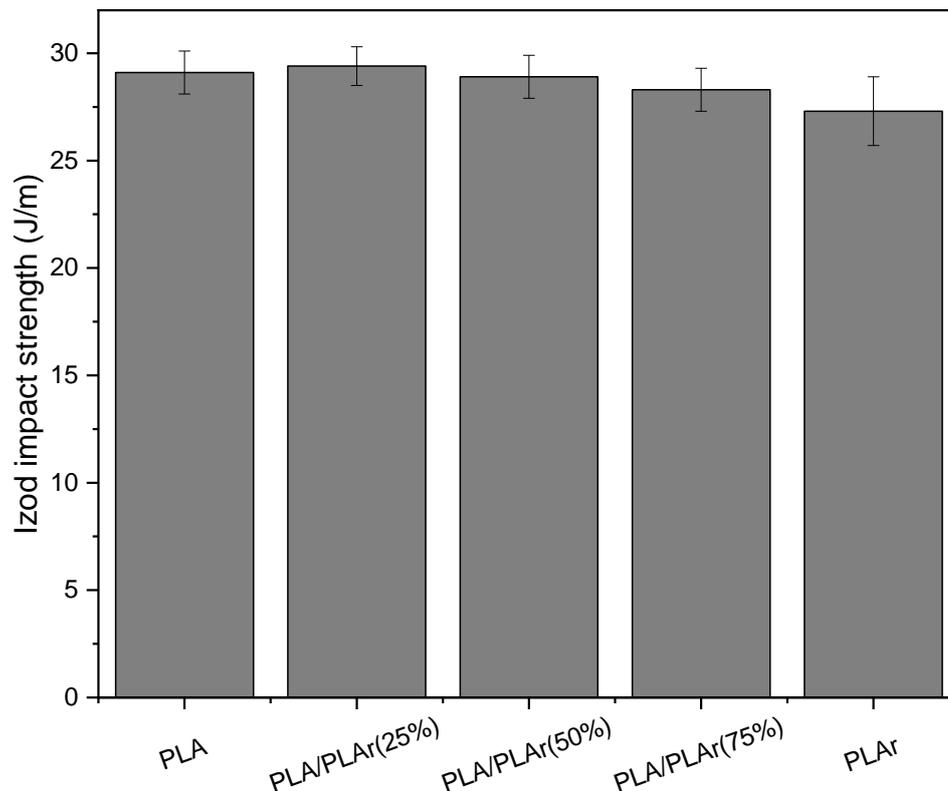


Source: Authors.

*Tensile Strength*

Figure 6 shows the results of the elastic modulus of neat PLA, PLAr and blends (PLA/PLAr), respectively. The PLA presented an elastic modulus around 1219.5 MPa, a typical value for fragile polymers (Barbosa et al., 2019). The elastic module of the PLAr was approximately 1139.3 MPa, corresponding to a slight decrease of 6.6%, in relation to the reference specimen of PLA. Thus, there is no significant difference in rigidity between neat PLA and PLAr, indicating the potential for reusing recycled resin. The addition of 25% PLAr to the blend (PLA/PLAr) did not influence the elastic modulus compared to neat PLA, only maintaining the value. Blends with 50% and 75% PLAr show a subtle reduction in the elastic modulus in relation to PLA, however this reduction is not significant. Although the PLAr has lower molar mass, as seen in the melt flow index, the elastic modulus of the blends was not severely affected. Such behavior is due to the elastic modulus being obtained in a very small deformation range (elastic region) and, consequently, it was not drastically affected.

**Figure 6**. Elastic modulus of PLA, PLAr and blends (PLA/PLAr).
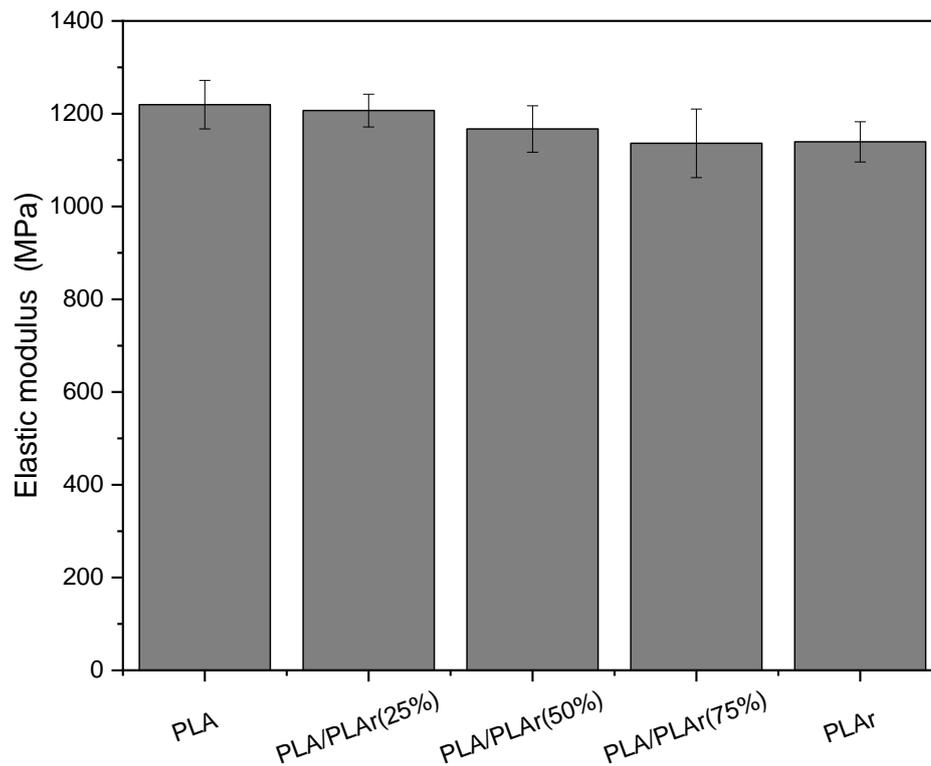


Source: Authors.

The tensile strength is generally related to the beginning of polymer molecule flow in the region of plastic deformation (Ferreira et al., 1997). Figure 7 shows the results of the tensile strength of neat PLA, PLAr and blends (PLA/PLAr), according to the content of recycled material, respectively. The PLA required a high tensile load to cause rupture, with a value around 60.3 MPa. On the other hand, PLAr fractured at lower stresses (47.8 MPa), indicating a reduction in the order of 20.7%, in relation to PLA. PLAr has a lower level of molecular entanglement, due to its lower molar mass, as seen in the melt flow index. As a consequence, there are fewer chains connected to the crystals to sustain the tension during the tensile test (Aurrekoetxea et al., 2001; Bahlouli et al., 2012), contributing to deteriorate the strength. The blends (PLA/PLAr) follow the rule of additivity since it can be noticed a continuous decrease in the tensile strength with the increase of the PLAr content.

**Figure 7**. Tensile strength of PLA, PLAr and blends (PLA/PLAr).
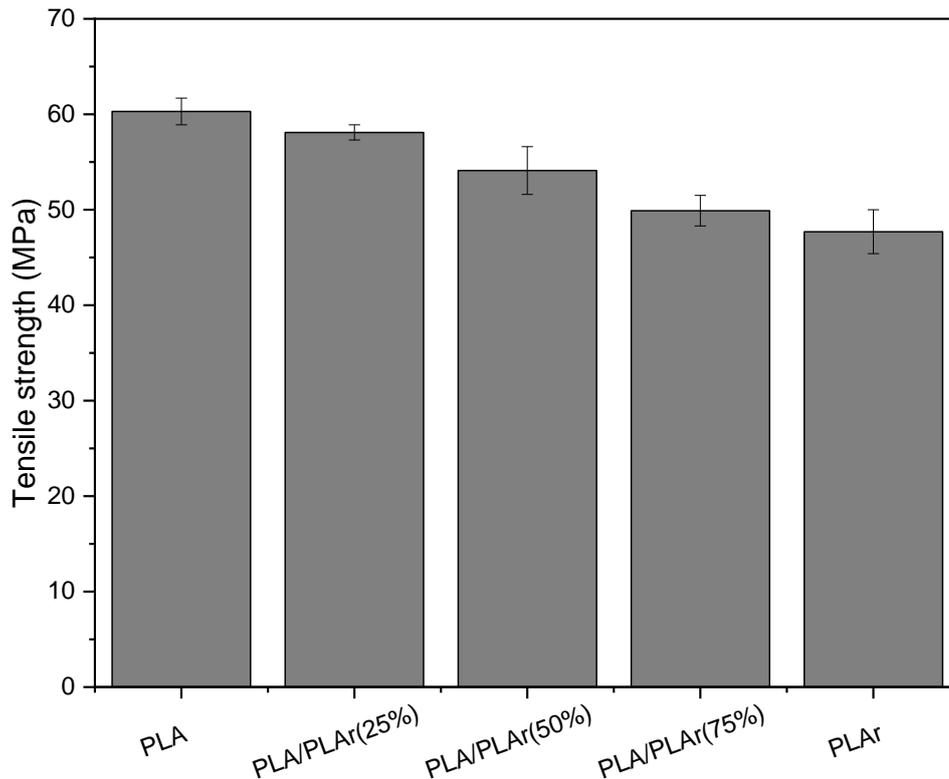


Source: Authors.

### *Shore D Hardness*

Figure 8 presents the Shore D hardness test results for neat PLA, PLAr and blends (PLA/PLAr), respectively. The PLA showed a Shore D hardness of around 77.9, while the lowest performance was found for the PLAr, with a value of 67.7 Shore D. The addition of 25% of PLAr in the neat PLA matrix caused a slight increase in Shore D hardness to 78.4, in relation to the PLA. When the PLAr concentration increased in the blends (PLA/PLAr) to 50% and 75%, there was a reduction in the resistance to penetration, with values of 74.4 and 69.1 Shore D, indicating that the surfaces became softer. Apparently, Shore D hardness property was governed by the degree of crystallinity, as verified in the DSC. The higher the degree of crystallinity, the higher the shore D hardness of the PLA/PLAr blends, suggesting greater wear resistance.

**Figure 8**. Shore D hardness of PLA, PLAr and blends (PLA/PLAr).
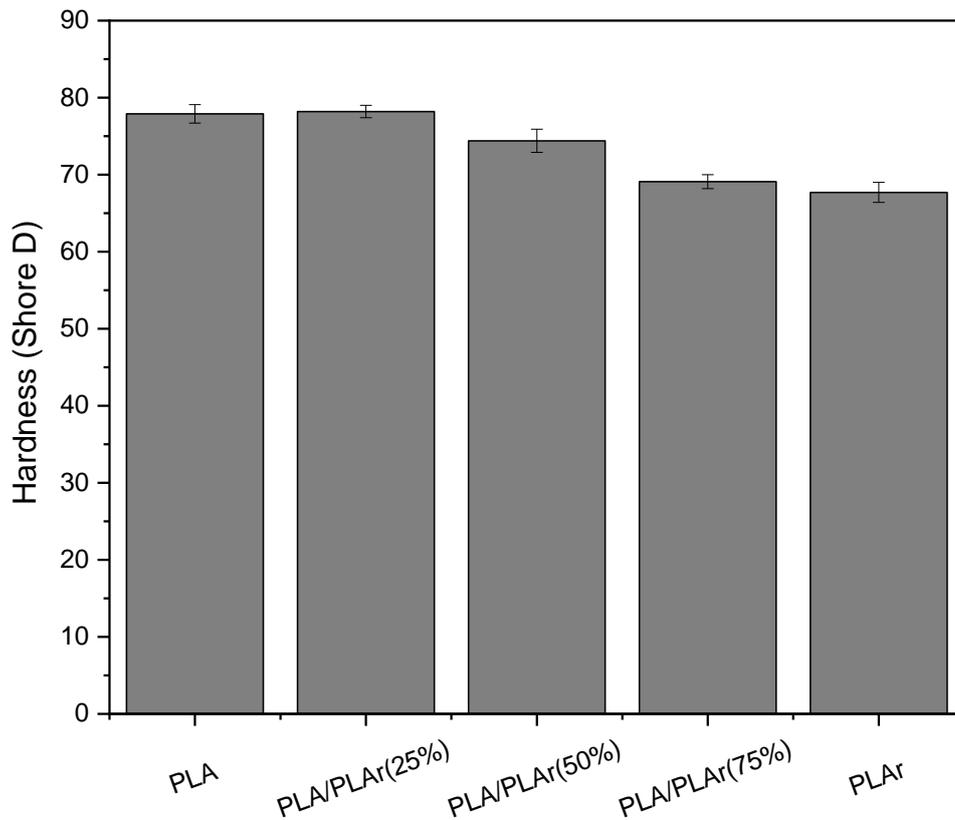


Source: Authors.

*** Heat Deflection Temperature (HDT)***

The structural stability of polymers at a temperature above the room's can be determined by means of the heat deflection temperature (HDT) (Rusayyis et al., 2018; Luna et al., 2019c). Figure 9 shows the results of the HDT for neat PLA, PLAr and blends (PLA/PLAr). PLA and PLAr showed HDTs in the order of 55.6 °C and 54.4 °C, respectively. In literature (Pereira and Morales, 2014), HDT values between 50-60 °C are reported for semicrystalline PLA. The addition of PLAr to the neat PLA, regardless of its content, did not significantly affect HDT, only a value maintenance. These results show that PLAr, even in high concentrations, did not damage the thermomechanical resistance of neat PLA, that is, PLA/PLAr blends present structural stability similar to neat PLA.

**Figure 9**. Heat deflection temperature (HDT) of neat PLA, PLAr and blends (PLA/PLAr).
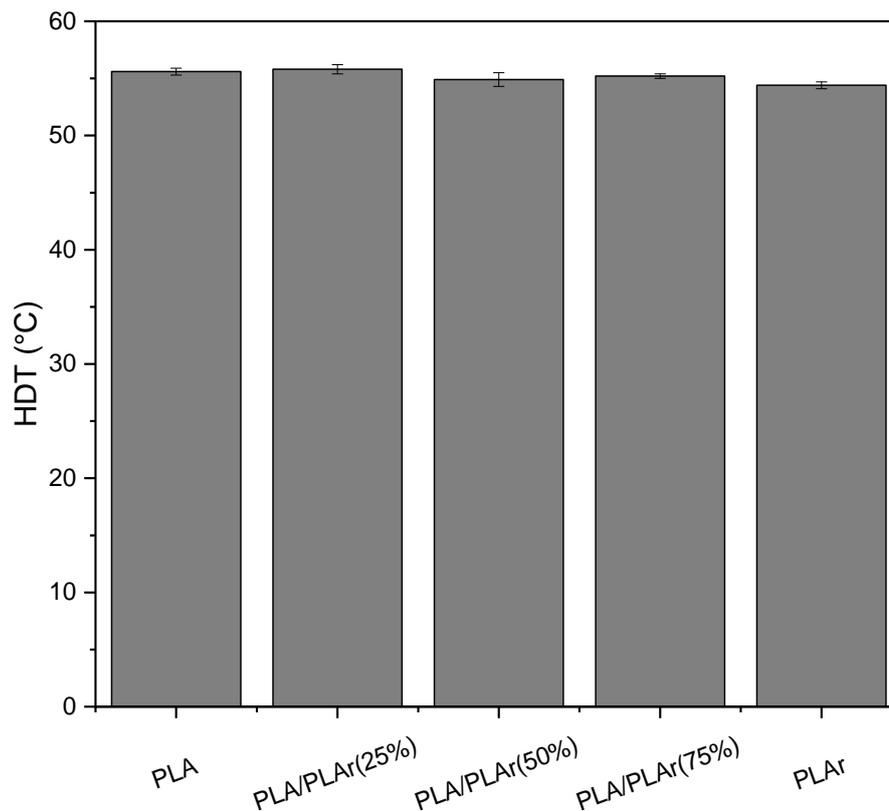


Source: Authors.

PLAr is a raw material with good properties and high added value, since it is not drastically compromising neat PLA. Although PLAr did not provide an increase in PLA's thermomechanical resistance, its presence maintained the structural stability of virgin resin. Therefore, there is a technological contribution to the development of ecological materials, such as PLA/PLAr blends.

*Vicat Softening Temperature (VST)*

Vicat temperature was used as a qualitative indicator of the system's thermomechanical behavior when subjected to a constant load and heating rate (Mélo et al., 2000; Ferreira et al., 2019b). Figure 10 shows the results of the Vicat softening temperature of neat PLA, PLAr and blends (PLA/PLAr). PLA had a Vicat softening temperature of 59 °C, a value within the range of the literature (Perego and Cella, 2010). PLAr and PLA/PLAr blends showed Vicat softening temperature at the same level as the neat PLA, indicating that thermomechanical stability was maintained, corroborating with the HDT analysis. The glass

transition temperature ($T_g$) values did not show significant variation in any case, with a range of 59-61 °C, as verified in the DSC. Note that the Vicat softening temperature values for PLA, PLAr and blends (PLA/PLAr) are found close to the $T_g$ range.

**Figure 10**. Vicat softening temperature of neat PLA, PLAr and blends (PLA/PLAr).
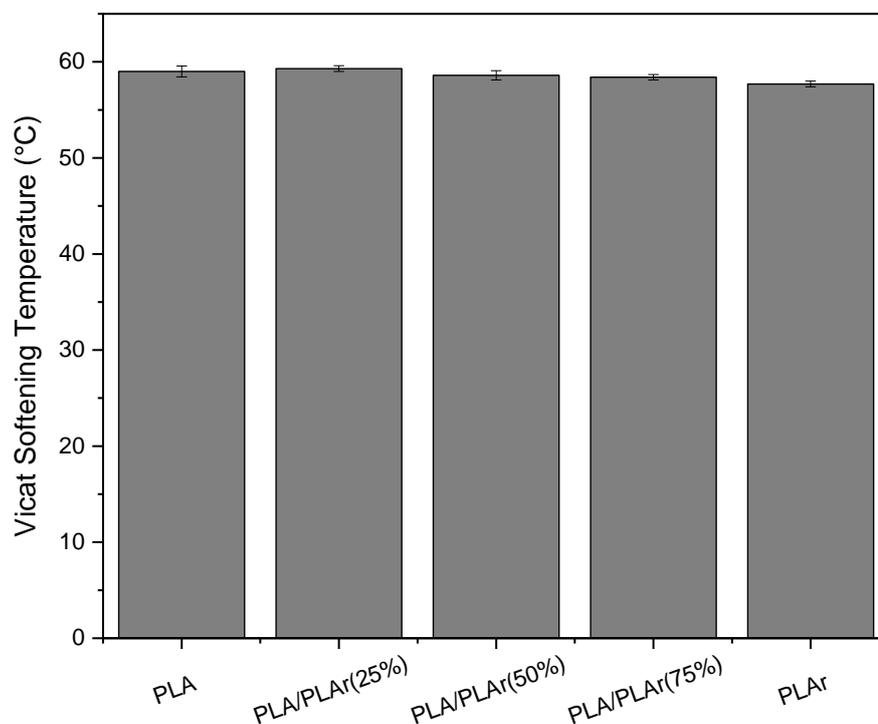


Source: Authors.

*Contact Angle*

Figure 11 presents the contact angle analysis results of neat PLA, PLAr and blends (PLA/PLAr) surfaces, respectively. The neat PLA had the highest contact angle (67.4°), while the PLAr had the lowest value (48.1°). Such behavior indicates that the neat PLA is more hydrophobic and the PLAr more hydrophilic, generating less and greater interaction with water, respectively. As PLAr contains pigments and, at the same time, it has already undergone primary mechanical recycling, it possibly increased the interaction with water. The literature (Cáceres et al., 2009; Luna et al., 2019d) has shown that degradation tends to reduce the contact angle, due to the surface becoming more polar. The PLA/PLAr blend (25%) showed a contact angle value close to the PLA, confirming the same trend of mechanical, thermomechanical, and thermal properties. However, there is a significant reduction in the

contact angle of the blends with 50% and 75% PLAr, indicating an increase in the degree of interaction with water. This finding suggests that from 50% of PLAr in the formulation of PLA/PLAr blends, there is a greater influence of the recycled material on the wetting behavior.

**Figure 11**. Images of water droplets on the surface of the materials after 200 seconds, for: (a) PLA; (b) PLA/PLAr (25%); (c) PLA/PLAr (50%); (d) PLA/PLAr (75%); (e) PLAr.

| $67.4 \pm 0.3$ | $63.8 \pm 0.4$ |
|---|---|



| $57.3 \pm 0.5$ | $50.2 \pm 0.4$ |
|---|---|



| $48.1 \pm 0.3$ |
|---|

**(e)**
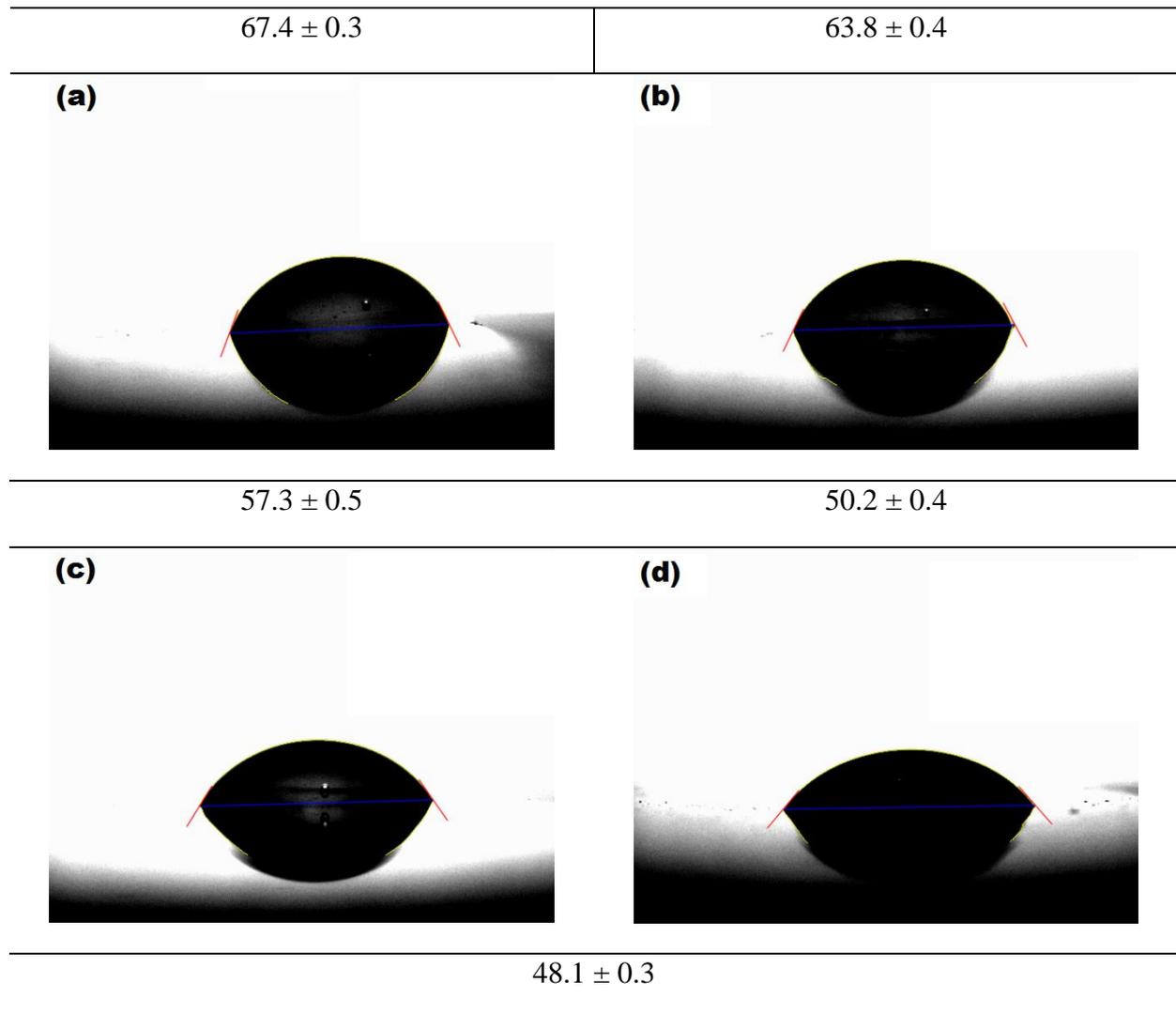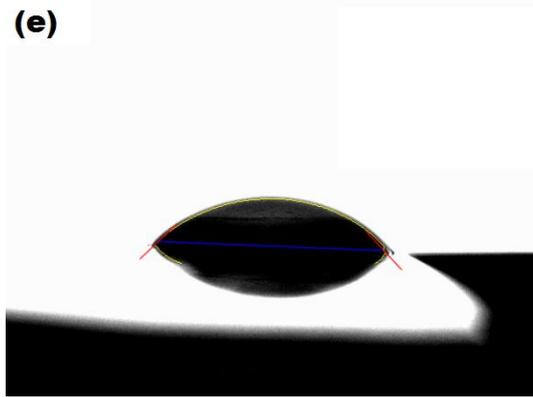
Source: Authors.

## 4. Conclusions

The technological potential of poly(lactic acid) (PLA) blends with 3D filament residues derived from poly(lactic acid) (PLAr) was investigated. The reuse of 3D filaments indicated a raw material with good properties, with feasibility to develop mixtures with neat PLA. In this case, one can replace part of the neat PLA resin with recycled one, generating materials with properties similar to PLA. When 25% of recycled PLA is used, there is a trend towards better thermal, mechanical, and thermomechanical properties, including staying on the same level as neat PLA. However, the use of 50% and 75% recycled PLA also showed potential for the development of blends, indicating that even high concentrations can be used in the formulations. In general, the present study showed that it is possible to manufacture products with good technical characteristics, using post-consumption material. In addition, the recycled PLA returns to the production chain, reducing the consumption of natural resources and energy, as well as contributing to sustainability.

The results also indicate a good prospect of application of these PLA waste. At the same time, it can minimize the harmful effect on the environment and add value to a discarded material.

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