Research Article

Eco-Friendly Approach and Potential Biodegradable Polymer Matrix for WPC Composite Materials in Outdoor Application

Alessia Quitadamo,[1,2] **Valerie Massardier,**[2] **and Marco Valente** ![ORCID][1]

[1]*Department of Chemical and Material Engineering, University of Rome La Sapienza, via Eudossiana 18, 00184 Rome, Italy*
[2]*Ingénierie des Matériaux Macromoléculaires, UMR CNRS 5223 Ingénierie des Matériaux Polymères, Bât J. Verne-INSA, 20, av Einstein, 69621 Villeurbanne Cedex Lyon, France*

Correspondence should be addressed to Marco Valente; marco.valente@uniroma1.it

Blends based on high-density polyethylene (HDPE) and poly(lactic) acid (PLA) with different ratios of both polymers were produced: a blend with equal amounts of HDPE and PLA, hence 50 wt.% each, proved to be a useful compromise, allowing a high amount of bioderived charge without this being too detrimental for mechanical properties and considering its possibility to biodegradation behaviour in outdoor application. In this way, an optimal blend suitable for producing a composite with cellulosic fillers is proposed. In the selected polymer blend, wood flour (WF) was added as a natural filler in the proportion of 20, 30, and 40 wt.%, considering as 100 the weight of the polymer blend matrix. There are two compatibilizers to modify both HDPE-PLA blend and wood-flour/polymer interfaces, i.e., polyethylene-grafted maleic anhydride and a random copolymer of ethylene and glycidyl methacrylate. The most suitable percentage of compatibilizer for HDPE-PLA blends appears to be 3 wt.%, which was selected also for use with wood flour. In order to evaluate properties of blends and composites tensile tests, scanning electron microscopy, differential scanning calorimetry, thermogravimetric analyses, and infrared spectroscopy have been performed. Wood flour seems to affect heavy blend behaviour in process production of material suggesting that future studies are needed to reduce defectiveness.

1. Introduction

The importance achieved by thermoplastic polymers during the last decades is undoubted. In fact, the thermoplastic global market is about 10% of the global chemical industry and displays one of the most important growths of the world economy [1]. Their development is strictly related to their mechanical properties, production process (easiness to perform serial production), cheapness, and versatile applications [2].

Unfortunately, thermoplastic diffusion did not correspond to an equal attention into end-of-life scenarios of these products. The consequence of this was an important pollution issue, significant on both earth and sea environment [3]. In this context, two strategies appear particularly suitable to be pursued: diffusion of bioderived polymers [4, 5] and production of composites with natural fillers [6, 7].

Polylactic acid (PLA) is one of the most diffused bioderived polymers on the market, thanks to its properties such as tensile strength, tensile modulus, and flexural strength, comparable or higher than other traditional polyolefins [8–10].

The main drawback of PLA is its high brittleness and limited toughness: to reduce it, many solutions have been suggested, such as the introduction of plasticizers, e.g., glycerol, oligomeric lactic acid, poly(ethylene glycol), and citrates [11]. These plasticizers are used in order to increase the intermolecular space between polymer chains, thanks to the introduction of small molecules, a procedure that results in their increased plasticity and resistance to shear. [12]. Another technique adopted in order to reduce PLA brittleness is producing blends with other thermoplastic polymers [13].

During the last decades, the production of polymer blends recognised as an important role in global markets as an easy method to tailor polymer properties to physical

TABLE 1: Formulations produced (10 samples for each family).

Samples	HDPE (%)	PLA (%)	Polybond 3029 (%)	Lotader AX8840 (%)	Wood flour (%)
HDPE50-PLA50	50	50			
HDPE50-PLA50-WF20	40	40			20
HDPE50-PLA50-WF30	35	35			30
HDPE50-PLA50-WF40	30	30			40
HDPE50-PLA50-Poly3-WF20	38.8	38.5	3		20
HDPE50-PLA50-Poly3-WF30	33.5	33.5	3		30
HDPE50-PLA50-Poly3-WF40	28.5	28.5	3		40
HDPE50-PLA50-Lot3-WF20	38.8	38.5		3	20
HDPE50-PLA50-Lot3-WF30	33.5	33.5		3	30
HDPE50-PLA50-Lot3-WF40	28.5	28.5		3	40

and mechanical requirements [14]. However, an effective mutual compatibility between polymers is necessary to obtain blends with higher properties than the original polymers. The development of an oil-based/bioderived thermoplastic blend is therefore the preliminary requirement of this work. In particular, an optimised high-density polyethylene and poly(lactic) acid blend could be produced in order to obtain oil-based/bioderived thermoplastic blends with high amount of bioderived charge, keeping good mechanical properties.

The second method, suggested to reduce plastic pollution, could be the development of thermoplastic matrix composites with natural fillers. Natural fillers gained great attention thanks to their appealing properties such as low density, low cost, renewability, recyclability, and biodegradability [15].

Among natural fillers, such as jute, flax, sisal, and paper cellulose fibres [16–21], wood flour exhibits interesting properties as waste material used for composite production, obtaining the well-known class of material wood plastic composites (WPC). WPC are thermoplastic matrix composites filled with wood-derived fillers [22–26], characterized by the potential recyclability and possibility to use waste as raw materials, reducing in this way environmental impact. It is widely recognised that a major issue with these composites is the not easy compatibility between hydrophobic polyolefin and hydrophilic natural fillers [27–30]. Many efforts have been done in order to face this problem, and the use of compatibilizers is one of the most diffuse. In fact, compatibilizers have a main hydrophobic chain compatible with thermoplastic polymer, while being grafted or added with copolymers in order to be compatible with hydrophilic charge.

The idea of this project is to develop oil-based/bioderived thermoplastics polymer blends as matrices for composites filled with natural fillers. In particular, we selected high-density polyethylene and poly(lactic acid) as, respectively, oil-based and bioderived polymers, while, as filler, we have chosen wood flour. Two kinds of compatibilizers have been tested in order to improve both blend properties and interfaces between blended matrix and wood flour: Polybond 3029 and Lotader AX8840 seem to be effective thanks to the presence of maleic anhydride grafted on polyethylene chains for the former and polyethylene random copolymer with glycidyl methacrylate for the latter. Moreover, an important purpose of this work is the valorisation of a natural waste, such as wood flour. As a consequence, the highest possible amount of wood flour has been tried to be added, compatibly with technological process limits.

2. Materials and Methods

Eraclene MP 90, commercial name of high-density polyethylene (HDPE) from ENI (Versalis), has been selected as oil-based polymer. Among its properties are a melt flow index (MFI) of 7 g/10 min (190°C/2.16 kg), a nominal mass of 0.96 g/cm^3, a tensile strength of 21 MPa, a tensile modulus of 1.2 GPa, and a Shore D hardness of 50.

Poly(lactic acid) (PLA) Ingeo Biopolymer 3251D from NatureWorks was selected as bioderived thermoplastic polymer, with a MFI of 35 g/10 min at 190°C/2.16 kg. This polymer is characterized by density 1.24, crystalline melting temperature in the range 155-170°C, and a glass transition temperature in the range 55-60°C.

Polybond 3029 has been selected as additive, suitable for cellulosic fillers. In fact, Polybond 3029 is a maleated polyethylene with a melt flow index of 4 g/10 min at 190°C/2.16 kg and the MA content is 1.7 wt.% (high). Generally, it is sold as pellets of 3-4 mm diameter.

Lotader AX8840 has been selected with the same purpose. It is a random copolymer of ethylene and glycidyl-methacrylate, with a melt flow index of 5 g/10 min at 190°C/2.16 kg. The GMA content is about 8 wt.%.

La.So.Le/est/Srl-Italy provided wood flour of hardwood beech as natural filler added to the PLA matrix. Wood flour has an average diameter of 250 μm.

Both PLA and wood flour were dried one night at 80°C, in order to avoid bubble formation due to water evaporation during the production process.

A Micro 15 Twin-screw DSM research extruder was used in order to produce samples. Temperature of 180°C, screw speed of 75 rpm, nitrogen atmosphere, and resident time of 4 min in the extruder have been selected to avoid PLA and wood flour degradation during the process.

Table 2: Tensile test results of samples filled with wood flour. Italic is for wood flour, bold for Polybond 3029, and underline for Lotader AX8840 addition.

| Samples	E (GPa)	σ (MPa)	$\mathcal{E}$ (%)
HDPE50-PLA50	1.88 ± 0.05	38.73 ± 0.18	99.38 ± 2.08
HDPE50-PLA50-WF20	*3.37 ± 0.14*	*34.60 ± 2.79*	*1.61 ± 0.01*
HDPE50-PLA50-WF30	*4.12 ± 0.11*	*34.00 ± 1.27*	*1.20 ± 0.20*
HDPE50-PLA50-WF40	*5.04 ± 0.17*	*33.50 ± 1.46*	*1.92 ± 0.03*
HDPE50-PLA50-Poly3-WF20	**3.30 ± 0.05**	**36.00 ± 0.83**	**1.81 ± 0.12**
HDPE50-PLA50-Poly3-WF30	**4.14 ± 0.20**	**35.90 ± 1.41**	**1.42 ± 0.10**
HDPE50-PLA50-Poly3-WF40	**4.95 ± 0.08**	**33.60 ± 1.75**	**0.93 ± 0.21**
<u>HDPE50-PLA50-Lot3-WF20</u>	<u>3.29 ± 0.04</u>	<u>37.80 ± 1.49</u>	<u>2.00 ± 0.11</u>
<u>HDPE50-PLA50-Lot3-WF30</u>	<u>4.06 ± 0.07</u>	<u>35.00 ± 1.19</u>	<u>1.42 ± 0.10</u>
<u>HDPE50-PLA50-Lot3-WF40</u>	<u>5.03 ± 0.12</u>	<u>34.10 ± 3.32</u>	<u>1.01 ± 0.13</u>
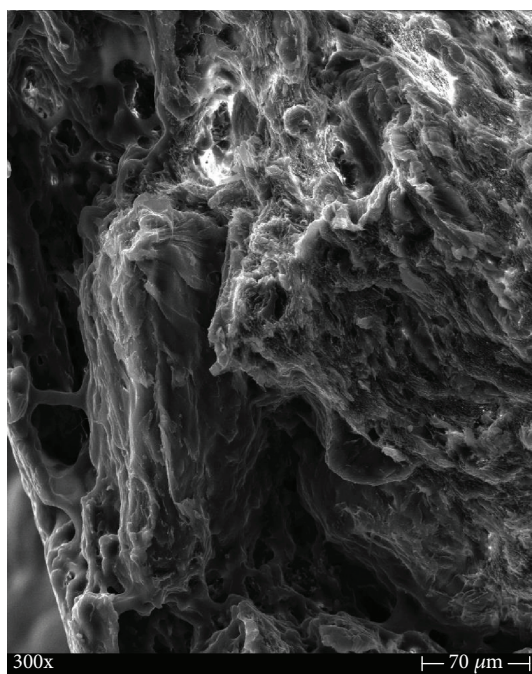
Figure 1: PE50-PLA50-WF30 composite.

Injection moulding has been used to obtain dog-bone specimens, with a mould temperature of 55°C and pressure parameters depending on polymer viscosity. For each family of samples, ten specimens have been produced. Table 1 sums up the formulations produced.

2.1. Tensile Tests. Tensile tests were performed in accordance with the ASTM D638 standard using ZwickRoell Z010, a load cell of 10 kN was used, and a 50 N preload was applied. A crosshead speed of 5 mm/min has been used. The tensile tests were performed on five dog-bone samples per series with a gauge length section $30 \times 4 \times 2$ mm^3 (L × W × T). For each family, five samples have been tested.

2.2. Scanning Electron Microscopy (SEM). Samples have been observed with Hitachi S2500 and 25 kV in order to analyse blend morphology and interfaces. Samples have been sputter-coated with gold particles before surface characterization.

2.3. Quartering. Samples produced, in the majority of cases, are characterized by high heterogeneity because of fibre dispersion and multiphase matrices. In order to obtain reliable results from thermal analysis and analyse a representative amount of sample, a cryogenic mill was adopted to obtain samples in the form of powders. A subsequent statistical approach, quartering, was used to select an exemplary amount of samples used for chemical and thermal analyses. This method is based on the separation of the total amount of charge in four parts equal in weight. Then, two parts at the opposite side are mixed together and the other two are separated.

2.4. Differential Scanning Calorimetry (DSC). Differential scanning calorimetry (DSC) tests were performed on a Q20 Thermal Analysis instrument from 25°C to 180°C at 10°C/min under a nitrogen flow of 50 mL·min^{-1}. Two cycles were performed with a 4-minute interval between them at 180°C to eliminate traces of thermal history. The first cycle provides information about properties after injection moulding, while the second one gives material properties. Cold crystallization, melting and crystallization parameters (temperature and enthalpy), and glass transition temperatures were analysed. ΔH_{mPE} (J/gPE) and ΔH_{mPLA} (J/gPLA) refer to enthalpy values to the exact amount of polyethylene and poly(lactic acid) in the samples.

2.5. Thermogravimetric Analysis (TGA). Thermogravimetric analysis (TGA) tests were carried out on a Q500 Thermal Analysis instrument, up to 600°C with a scanning temperature of 10°C/min under a nitrogen flow of 50 mL min^{-1}. From these analyses, we derived temperatures at which degradation started (T_{onset}), evaluated through the
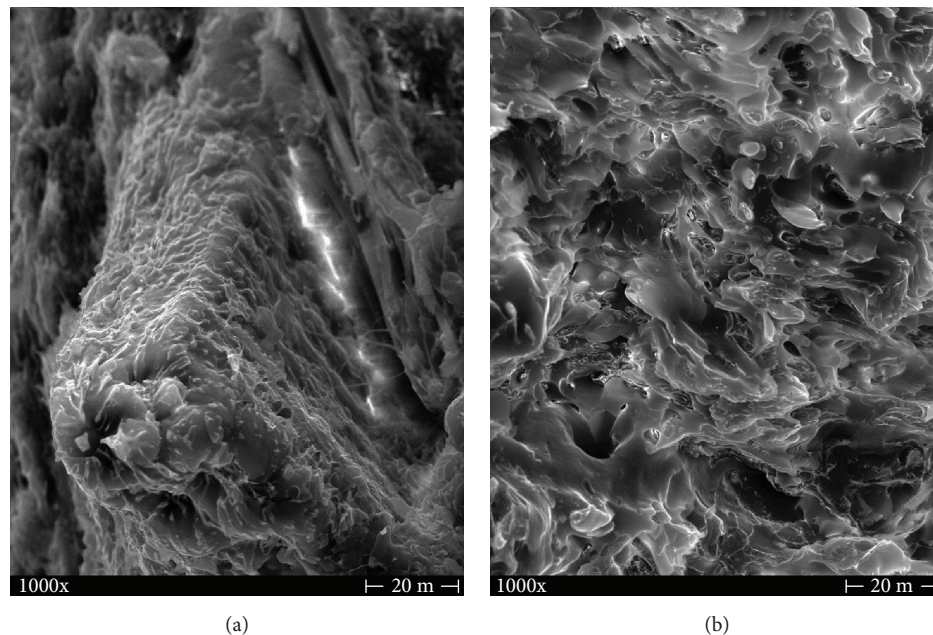
(a)

(b)

FIGURE 2: PE50-PLA50-Lot3-WF30 (a) and PE50-PLA50-Poly3-WF30 (b).

TABLE 3: DSC results for HDPE50-PLA50 composites with 20, 30, and 40 wt.% of wood flour and 3 wt.% of compatibilizers Polybond 3029 and Lotader AX8840.

	ΔH_{ccPLA} (J/g_{PLA})	T_{cc} (°C)	ΔH_{mPE} (J/g_{PE})	T_{mPE} (°C)	ΔH_{mPLA} (J/g_{PLA})	T_{mPLA} (°C)	T_{gPLA} (°C)	X_{HDPE} (%)	X_{PLA} (%)
HDPE	—	—	215	134	—	—	—	73	—
PLA	7	98	—	—	41	168	61	—	44
HDPE50-PLA50	8	97	236	132	40	168	62	81	34
HDPE50-50PLA50-WF20	—	—	190	132	35	167	61	65	38
HDPE50-50PLA50-WF30	—	—	200	132	39	167	62	68	42
HDPE50-50PLA50-WF40	—	—	193	132	40	167	63	66	43
HDPE50-PLA50-Poly3-WF20	15	99	202	132	45	167	61	69	32
HDPE50-PLA50-Poly3-WF30	16	99	186	132	45	167	61	63	31
HDPE50-PLA50-Poly3-WF40	9	97	187	132	42	165	59	64	35
HDPE50-PLA50-Lot3-WF20	6	100	187	132	44	167	61	64	35
HDPE50-PLA50-Lot3-WF30	6	99	189	132	45	166	60	65	42
HDPE50-PLA50-Lot3-WF40	3	98	184	132	42	166	61	63	42

extrapolated onset temperature from TGA curve, and Δm, the mass variation during the test.

2.6. Infrared Analysis Attenuated Total Reflection (ATR-FTIR). Infrared analysis attenuated total reflection (ATR-FTIR) tests were carried out with a Thermo Scientific Nicolet iS10 spectrometer, with a spectral range 4000-400 cm^{-1} and 32 scans.

3. Results and Discussion

3.1. Tensile Tests. Mechanical properties of these composites have been evaluated to detect the influence of wood flour on blend properties. In another study, we analysed the influence of compatibilizers on HDPE-PLA blends. In particular, we studied the effect of 1.3 and 5 wt.% of Lotader AX8840 and

Polybond 3029 on HDPE-PLA compatibility. The optimal compatibilizer percentage has been selected as 3 wt.% in both cases. We evaluated, as a consequence, the effect of 3 wt.% of both Lotader AX8840 and Polybond 3029 on HDPE50-PLA50-WF composites.

Lotader AX8840 and Polybond 3029 were selected because of their ability to interact with both polyethylene and polymers with polar groups. Moreover, Lotader AX8840 and Polybond 3029 could interact with natural fillers, such as wood flour. Wood flour was only added on samples HDPE50-PLA50-Poly3 and HDPE50-PLA50-Lot3 in three different percentages (20, 30, and 40 wt.%). In fact, higher wood flour percentages have not been added because of processing technological limits. Table 2 sums up tensile test results for these families of samples.
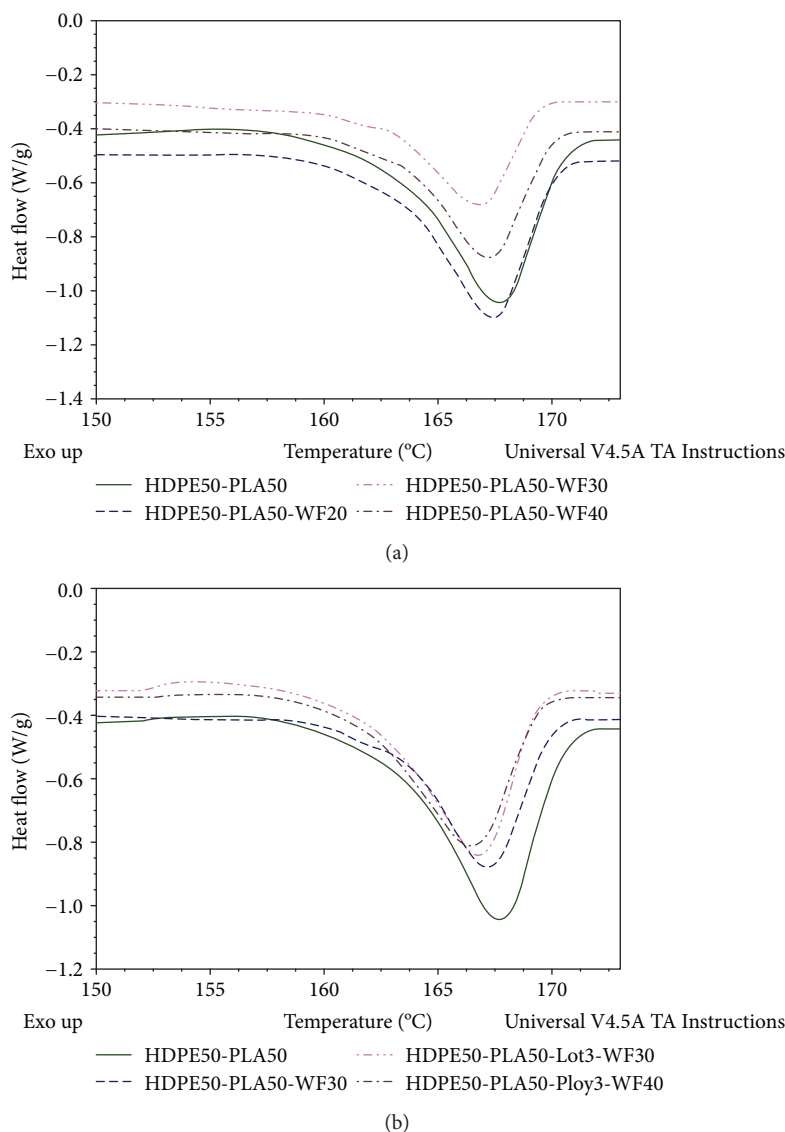
(a)

(b)

FIGURE 3: (a) PLA melting peak magnification of HDPE50-PLA50, HDPE50-PLA50-WF20, HDPE50-PLA50-WF30, and HDPE50-PLA50-WF40. (b) PLA melting peak magnification of HDPE50-PLA50, HDPE50-PLA50-WF30, HDPE50-PLA50-Lot3-WF30, and HDPE50-PLA50-Poly3-WF40.

Wood flour addition improved elastic modulus at the strong expense of elongation at break: increased wood flour percentages resulted in higher values of the elastic modulus, yet tensile strength did not experience the same growth. The latter result was attributed to poor interface between HDPE50-PLA50 matrix and wood flour (Figure 1) [31].

The addition of Polybond 3029 and Lotader AX8840 in the amount of 3 wt.% did not strongly affect mechanical properties of the composite; all the results are comparable to HDPE50-PLA50-WF for all three wood flour percentages. Probably, the presence of 3 wt.% did not prove sufficient to have a significant effect on the strength of both HDPE-PLA and polymer blend-wood flour interfaces. One could suppose that the compatibilizer's amount of 3 wt.% slightly acts on blend matrix-wood flour interfaces, but it was not sufficient to satisfy the total improvement of the composites.

Future studies would need to concentrate on optimizing the amount of compatibilizers, or to analyse chemical treatments on wood flour that could improve blend matrix-wood flour interfaces.

3.2. Scanning Electron Microscopy (SEM). Scanning electron microscopy (SEM) analyses are aimed at investigating the soundness of interfaces between blended matrix and wood flour. HDPE50-PLA50 morphology has been studied in another study. In particular, a typical immiscible blend morphology has been displayed by HDPE and PLA. The addition of compatibilizers, especially Lotader AX8840, seems to increase the compatibility between polymers.

Wood flour addition increased morphology complexity, in agreement with mechanical properties, especially low elongation at break. Wood particles seem to be partially

TABLE 4: TGA results for HDPE-PLA blends with different compatibilizer percentages and HDPE50-PLA50 matrix composites with 3 wt.% of compatibilizer and 20, 30, and 40 wt.% of wood flour. T_{onset} (°C) is evaluated with the extrapolated onset temperature from TGA curve. T_{DTG} (°C) is the temperature of maximum DTG curve peaks. Δm (%) is the mass variation percentage between the total amount of sample before the test and the residual mass after the test.

	T_{onset} (°C)	T_{DTG} (°C)	Δm (%)
HDPE	458	474	100
PLA	319	351	100
Poly	459	480	100
Lot	434	464	100
WF	289	292/349	74
HDPE50-PLA50	322	345/471	100
HDPE50-50PLA50-WF20	294	315/465	95
HDPE50-50PLA50-WF30	287	313/469	93
HDPE50-50PLA50-WF40	284	314/469	91
HDPE50-PLA50-Poly3-WF20	309	347/472	95
HDPE50-PLA50-Poly3-WF30	302	335/472	93
HDPE50-PLA50-Poly3-WF40	291	332/470	91
HDPE50-PLA50-Lot3-WF20	296	324/472	95
HDPE50-PLA50-Lot3-WF30	286	320/470	93
HDPE50-PLA50-Lot3-WF40	285	322/472	91

related to the polymer matrix, but not completely compatibilized. In particular, as suggested from other tests, we could suppose that PLA has more affinity for wood flour than HDPE. However, a high inhomogeneity of the blended matrix is displayed (Figure 1).

The presence of Lotader AX8840, thanks to glycidyl methacrylate groups, seems to improve matrix homogeneity, increasing at the same time the interface with wood flour (Figure 2(a)). The presence of Polybond 3029 did not allow obtaining a sensitive reduction in matrix defectiveness: in contrast, phase separation is still clearly visible (Figure 2(b)). In fact, in a previous study [32], we analysed blends of PLA with both Lotader AX8840 and Polybond 3029. Higher homogeneity has been displayed by a PLA-Lotader AX8840 blend, probably because of a higher glycidyl methacrylate content (about 8 wt.%) compared to maleic anhydride content in Polybond 3029 (1.5-1.7 wt.%).

3.3. Differential Scanning Calorimetry (DSC). Differential scanning calorimetry has been performed to evaluate the influence of compatibilizers and wood flour on HDPE and PLA.

In order to analyse wood flour effect on HDPE-PLA blends (compatibilized or not), Table 3 displays DSC results. PLA particles dispersed in the HDPE matrix seems to act as a nucleating agent on HDPE, increasing its crystallinity in HDPE50-PLA50 [33]. The typical PLA cold crystallization phenomena are not revealed with wood flour addition, suggesting a higher facility of PLA macromolecules to arrange during cooling from molten state. In fact, we can suppose a nucleating effect of wood flour on the PLA phase,

increasing its crystallinity. Moreover, the higher wood flour amount in HDPE50-PLA50-WF composites, the lower the HDPE crystallinity, suggesting a lower PLA-nucleating effect on HDPE. Both Polybond 3029 and Lotader AX8840 added to HDPE-PLA-WF composites display a positive effect in terms of compatibility: a slight reduction in PLA glass transition temperature was evident with respect to composites without compatibilizers. HDPE and PLA crystallinity have been evaluated as

$$X(\%) = \frac{\Delta H_m}{\Delta H_t} \times \frac{1}{w} \times 100, \qquad (1)$$

where ΔH_m is the experimental enthalpy value from DSC analysis, ΔH_t is the theoretical melting enthalpy value of fully crystalline HDPE (293 J/g) [10] or fully crystalline PLA (93 J/g) [11], and w is the weight fraction of polymer in the composites. If a cold crystallization phenomenon occurs, $X(\%)$ has to be evaluated as

$$X(\%) = \frac{\Delta H_m - \Delta H_{cc}}{\Delta H_t} \times \frac{1}{w} \times 100. \qquad (2)$$

Another compatibility influence of Lotader AX8840 and Polybond 3029 can be extrapolated by analysing PLA crystallization. In particular, Polybond 3029 seems to have a higher effect on compatibility between wood flour and PLA, obtaining lower crystallinity of PLA and, as a consequence, higher cold crystallization enthalpy. In fact, generally speaking, the higher the compatibility, the lower the nucleating effect [33].

For HDPE50-PLA50-WF composites, DSC thermograms (Figure 3) also reveal the presence of a shoulder for PLA melting peak, suggesting the presence of two different crystalline phases, in agreement with an effect of WF on PLA crystallinity. Similarly, this effect has been displayed with talc addition in PLA-based composites [34]. In fact, a double melting peak is related to two different crystalline phases.

3.4. Thermogravimetric Analysis (TGA). Studying the mass variation of samples Δm (%), only samples filled with wood flour display a residual char after the test (Table 4), as a result of wood pyrolysis. As a consequence, the higher the amounts of wood flour, the higher the residual mass. In general, firstly evaporation of adsorbed moisture occurs from room temperature to 100°C. Secondly, degradation of hemicellulose occurs around 295°C, while cellulose pyrolysis occurs at a higher temperature (315-400°C) [35]. PLA onset degradation temperature is around 319°C, near the degradation of WF. In fact, HDPE50-PLA50-WF composites display a unique degradation peak for PLA and WF degradation [36], while a second peak, at higher temperatures, is displayed for HDPE degradation around 470°C.

The addition of Lotader AX8840 seems to increase the thermal stability of blended matrix composites. A possible hypothesis would be that PE-based compatibilizers interact with PLA and wood flour: maleated groups seem to be more efficient with the presence of wood flour, increasing
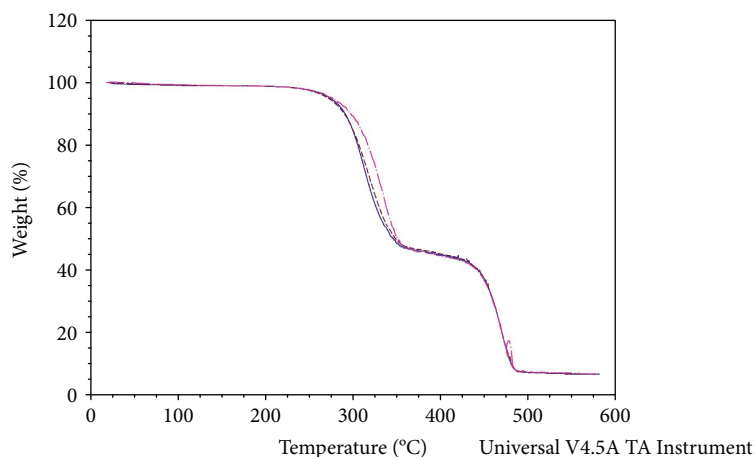
FIGURE 4: Mass variation as a function of temperature during TGA analyses of HDPE50-PLA50-WF40 (blue), HDPE50-PLA50-Lot3-WF30 (red), and HDPE50-PLA50-Poly3-WF30 (purple).
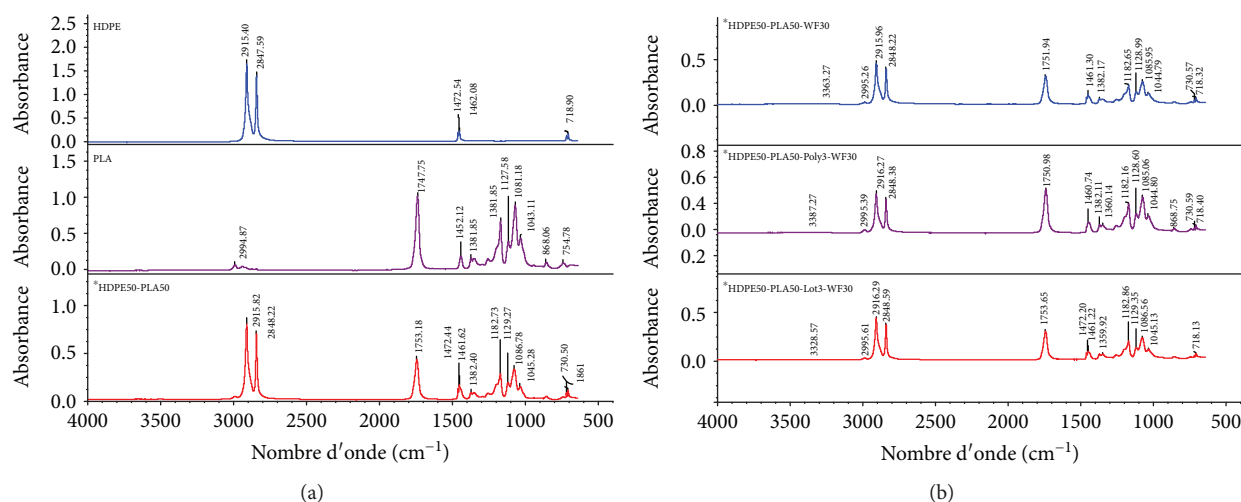

(a)

(b)

FIGURE 5: ATR-FTIR results for HDPE-PLA blends (a) and HDPE50-PLA50 matrix composites with 3 wt.% of compatibilizer and 30 wt.% of wood flour (b).

the thermal stability of blended matrix composites, while the presence of glycidyl methacrylate seems to be more efficient with PLA, increasing the thermal stability of the HDPE-PLA blend. We already propose a higher affinity between PLA and Lotader AX8840 in a previous work [31], displaying a smaller secondary phase for Lotader AX8840 than for Polybond 3029 when blended with PLA. In fact, as visible from Figure 4, a shift of PLA and WF degradation step is displayed when compatibilizers are added, in agreement with the interaction between biobased products and compatibilizers.

3.5. Infrared Analysis Attenuated Total Reflection (ATR-FTIR). Infrared spectroscopy has been done to evaluate the interactions between polymers and wood flour, and the main results are displayed in Figure 5. As expected, HDPE50-PLA50 did not display peak variation (Figure 5(a)) confirming the presence of an immiscible blend without interactions between HDPE and PLA [37]. A slight interaction was displayed by wood flour with the HDPE50-PLA50 matrix, especially with

PLA. In fact, a little shift of a typical C=O peak of PLA ($1749\,cm^{-1}$) and OH peak of wood flour ($3334\,cm^{-1}$) was displayed by HDPE50-PLA50-WF20, HDPE50-PLA50-WF30, and HDPE50-PLA50-WF40. A shift variation of the OH peak in blended matrix composites with compatibilizers, both Polybond 3029 and Lotader AX8840, also suggest interactions (Figure 5(b)).

4. Conclusions

Previously, a good balance of properties and amount of bio-derived polymer has been obtained for HDPE-PLA blends, in which the amounts of HDPE and PLA were the same. The addition of wood flour allows a further reduction in the content of oil-based polymer, allowing, at the same time, an improvement in mechanical properties. Unfortunately, wood flour addition also increases matrix defectiveness. The addition of compatibilizers slightly improves matrix homogeneity and interaction with wood flour, modifying at the same time thermochemical properties of the

composites. Future studies will concern the investigation of optimal compatibilizer percentage in order to increase both HDPE-PLA compatibility and blended matrix-wood flour interfaces. Moreover, a higher natural filler percentage can be tested, trying also different morphology interactions such as wood flour and recycled paper fibres. To conclude analyses, biodegradability tests have to be carried out [38], in order to correlate mechanical-thermal properties with end-of-life behaviour of composites. Moreover, infrared thermography analyses will be performed, to analyse the compatibility between natural fillers (such as wood flour and recycled paper fibres) and the polymer matrix.

Data Availability

Previously reported data used to support this study are available at doi:10.1016/j.compositesb.2016.11.013, doi:10.1002/pen.24606, and doi:10.1515/hf-2017-0149. These prior studies are cited at relevant places within the text as references [21, 26, 31].

Conflicts of Interest

The authors declare that they have no conflicts of interest regarding the publication of this paper.

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