

## Article

# Mechanical Response of Reactive Extruded Biocomposites Based on Recycled Poly(lactic Acid) (R-PLA)/Recycled Polycarbonate (R-PC) and Cellulosic Fibers with Different Aspect Ratios

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**Abstract:** Coupling recycling processes with increased use of bio-derived and environmentally friendly materials, with the aim of approaching (or overcoming) the mechanical properties of petroleum-derived plastics, is a path that research is pursuing in small but important steps. It is in this stream that this paper wants to fit in developing recycled poly(lactic acid) (R-PLA)/recycled polycarbonate (R-PC) blends obtained from thermoforming processing scraps and reinforcing them with cellulosic-derived fibers, having three different aspect ratios. The aim is to understand the mechanical properties of “second life” materials, their adherence to some micromechanical predictive models and the reinforcement capacity of these natural fibers in relation to their dimensions. Moreover, a compatibilizing system, based on Triacetin (TA) and Tetrabutylammonium Tetraphenylborate (TBATPB), has been added during the extrusion to investigate if a reactive process among R-PLA/R-PC and cellulosic fibers can be achieved.

**Keywords:** biocomposites; reactive extrusion; recycled polymers; mechanical behavior



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## 1. Introduction

Nowadays, in the field of materials, a stringent goal of both academia and industry is the optimization of raw material consumption efficiency to minimize waste [1]. For this purpose, the use of materials of bio-based origin and/or derived from recycling is a tough necessity [2]. In relation to durable applications, the use of these polymers must be considered in relation to the “performance” that the finished products must possess (to grant a significantly long second life), coupled with an easy type of processing [3,4]. The current industrial chain, going from formulation design, compounding up to the final shaping, should not be possibly altered in terms of possibly not altering the existing types of machinery, energy consumption and time, to avoid industry-prohibitive investments [5].

The introduction of plastics in a productive process must meet regulatory requirements already at the design projecting stage, where materials, manufacturing and design are defined according to the effects of the design on material recovery after the end-of-life treatment [6] and by avoiding the presence of contaminants without losing performances in the future recycled product [7]. Recycled plastics can replace oil-derived polymers in several applications and offer benefits from a production and economic point of view [8,9]. There are, however, still significant limitations in reprocessed plastics due to lack of performance [10] that makes their use tricky as potential matrices for sustainable biocomposites [11].

In this framework, eco-composites characterized by natural fibers and recycled matrices are considered very attractive due to the combination of appealing mechanical and physical properties and high sustainability [12]. The trend toward the use and development of eco-friendly raw materials will be even more dominant in future years since regulations

are promoting this concept; for example, in the automotive field, it will be mandatory that 85% of plastic components of a vehicle must come from a recycling process [13].

In this context, the idea of the present study is to develop short-fiber biocomposites starting from poly (lactic acid) (PLA)/polycarbonate (PC) blends obtained from recycled materials (derived from processing scraps), reinforcing them with cellulosic fibers, having three different aspect ratios and being confident that they can be processed at the temperatures and extrusion times required having tested their thermogravimetry in previous studies [14,15].

Poly (lactic acid) has interesting physical and mechanical properties (high stiffness, good processability and industrial biodegradability) [16,17], but the requirements of durable applications are very demanding. PLA has, indeed, low thermal stability [18], low toughness (Charpy Impact Strength around 3 kJ/m<sup>2</sup> [19]) and low tensile elongation [20]. It is, therefore, necessary to improve these properties to use PLA in long-term and large-scale applications. In order to improve the thermal stability of PLA-based materials, the strategy of blending it with bisphenol A poly(carbonate) (PC), having a higher glass transition temperature [21], can be a viable solution.

It is pivotal to consider that the processability window of PC involves temperatures of around 300 °C [22] and several companies produce polymer blends (as PC/ABS (Bayblend, Lexan) [23] or PC/PBT (Markoblend, Xenoy) [24] that allow processing at lower temperatures. PLA can be able to replace ABS; this not only results in modulated properties but also reduces the environmental impact of the materials [25]. Nevertheless, PLA/PC blends are immiscible, and the adhesion between the two polymers is weak due to the high surface tension; therefore, a correct compatibility method must be selected [26].

A route pursued to improve PLA/PC adhesion was the study of the effect of various catalysts on the interchange reactions of PLA and PC [27]. These attempts were made to clarify the reaction mechanism by preparing mixtures in a batch mixer and adopting a mixing time of 1000 s. The result was surprising; in fact, the presence of PLA-PC copolymer was detected by means of thermal-dynamic-mechanical analysis (DMTA): a new glass transition temperature intermediate between those of pure PLA and PC was identified. This methodology has also been followed by other researchers [28,29] with similar results. To achieve the desired compatibility in a shorter time, more in line with the residence time of a typical twin-screw extruder, Phuong et al. [30] proposed a study in which both Triacetin (TA) and tetrabutylammonium tetraphenylborate (TBATPB) were added to PLA/PC blends for a short mixing time of 60 s. TA is an excellent humectant plasticizer, and thanks to its high mobility in the melt, promotes more effective interchange reactions between PLA and PC catalyzed by TBATPB [31]. The occurrence of these reactions was demonstrated by an improvement of the mechanical and thermal properties of the PLA/PC compatibilized blends as a function of the composition [32]. Moreover, Phuong et al. [33] also prepared biocomposites by melt extrusion based on virgin PLA/PC blends improving the interaction between natural fibers and polymer matrix as demonstrated by selective extraction of polymers and infrared analysis of the obtained residual fraction, showing weak stretching vibration of ester groups chemically bonded to microcellulose fibers.

The step forward of the present work is to study such systems composites consisting of the blend based on recycled PLA and recycled PC reinforced, with microcellulose fibers having three different aspect ratios but with a matrix consisting of a recycled PLA/PC blend. The aim is to understand the variation in the mechanical properties of these materials after a life cycle and to explore with a modeling approach the fitting of some predictive models to experimental data. Moreover, the optimized catalyst system, always based on TA and TPB, has been used during melt extrusion to investigate if a better adhesion between the components can be achieved in its effectiveness as an interfacial adhesion enhancer in this material.

## 2. Materials and Methods

### 2.1. Materials and Compositions

- R-PLA: It is an industrial processing waste recovered from products obtained through a thermoforming process with PLA2003D (Natureworks LLC, Minnetonka, MN, USA), a thermoplastic resin with a density of 1.24 g/cm<sup>3</sup>, extrusion grade. The regrinding of the scraps (previously passed through a metal detector to avoid contamination of the material) has been achieved by Romei s.r.l (Florence, Italy), and it is followed by further filtering and homogenization of the granules before subsequent use.
- R-PC: The same process explained in the first bullet point, an example of post-industrial recovery becoming raw material, is used by Romei s.r.l. (Firenze, Italy) to recover, regrind and sieve recycled polycarbonate scraps from thermoformed objects produced with Bisphenol A Polycarbonate grade S3000 (Mitsubishi Chemical Co., Tokyo, Japan) having a density of 1.20 g/cm<sup>3</sup>.
- CATA: Triacetin (TA), also known as glycerine triacetate, and TetraButylAmmonium TetraPhenylBorate (TBATPB), both purchased from Sigma-Aldrich—Merck, were used as catalysts for interchange reactions. Phuong et al. [32] explained how the reaction occurs from a mechanistic point of view. The interchange reaction of the modified PLA with PC can produce block copolymers with varying lengths and branching points. The presence of branching creates an irregular structure that is probably responsible for the peculiar thermal properties of the blends. In the presence of cellulose fibers, the catalyst will also interact with it due to the hydroxyl groups present on the surface of the cellulose itself.
- Lyocell fibers: Tencel<sup>®</sup> FCP-10/400 microfibers (Lenzing, Austria) with an aspect ratio of 36 were provided by Lenzing AG, Austria. According to this manufacturer, these fibers have a density of 1.5 g/cm<sup>3</sup>.
- Arbocel fibers: Two different types of extra short-fibers, kindly provided by J Rettenmaier Sohne<sup>®</sup> (Rosenberg, Germany), were used:
  - Arb I (ARBOCEL<sup>®</sup> 600BE/PU) (mean diameter 20 μm, mean fiber length 60 μm, mean aspect ratio 3, bulk density: 200–260 g/L).
  - Arb II (ARBOCEL<sup>®</sup> BWW40) (mean diameter 20 μm, mean fiber length 200 μm, mean aspect ratio 10, bulk density: 110–145 g/L).

To carry out a systematic comparative study of a biocomposite that was not only completely obtained from recyclable products and natural fibers but also had a predominantly biobased matrix (PLA), the following formulations reported in Table 1 (comparing systems with and without CATA) were produced and characterized.

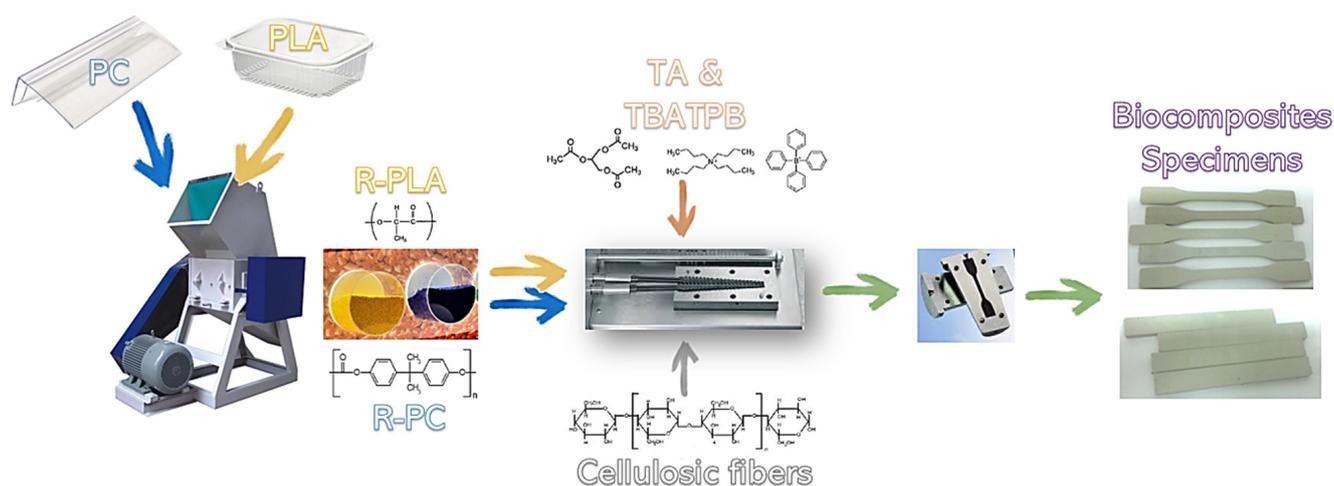
**Table 1.** Biocomposites composition.

Formulation	R-PLA (wt. %)	R-PC (wt. %)	Arb I (wt. %)	Arb II (wt. %)	Lyocell (wt. %)	TA (wt. %)	TBATPB (wt. %)
R-PLA60/R-PC40 + 10ArbI	54	36	10	-	-	-	-
R-PLA60/R-PC40 + 10ArbII	54	36	-	10	-	-	-
R-PLA60/R-PC40 + 10Lyo	54	36	-	-	10	-	-
R-PLA60/R-PC40 + 10ArbI + CATA	50.9	33.9	10	-	-	5	0.2
R-PLA60/R-PC40 + 10ArbII + CATA	50.9	33.9	-	10	-	5	0.2
R-PLA60/R-PC40 + 10Lyo + CATA	50.9	33.9	-	-	10	5	0.2

### 2.2. Processing

After carrying out the drying of the material in a vacuum oven at 60 °C for 24 h, the materials were processed to obtain biocomposites using a micro twin-screw extruder (Thermo Scientific HAAKE MiniLab II, Karlsruhe, Germany) capable of processing 5 g of material. The process temperature was set at 235 °C for 60 s in order to avoid PLA degradation but to ensure the complete melting of the blend, its mixing, the homogeneous dispersion of the fibers and the achievement of the catalytic process.

The exiting melt is connected, via a heated cylinder piston, with a mini-injection molding machine (Thermo Scientific HAAKE Minijet II, Karlsruhe, Germany), which allows the molding of specimens with defined geometry. The mold was held at 60 °C and the molding cycle time was around 25 s. The processing route is shown in Figure 1.



**Figure 1.** Processing route to obtain biocomposite samples.

### 2.3. Testing Methodologies

Tensile tests were carried out at room temperature on Haake type III dog-bone tensile bars (size: 25 × 5 × 1.5 mm) at a crosshead speed of 10 mm/min by an Instron 5500R universal testing machine (Canton, MA, USA), equipped with a 10 kN load cell and interfaced with a computer running MERLIN software (INSTRON version 4.42 S/N-14733H).

Impact tests were performed on V-notched 80 mm × 10 mm × 4 mm specimens using a 15 J Instron CEAST 9050 Charpy pendulum (INSTRON, Canton, MA, USA) following the standard procedure ISO 179.

Dynamic Mechanical Thermal Analysis (DMTA) was performed on a Gabo Eplexor<sup>®</sup> 100N (Gabo Qualimeter GmbH, Ahlden, Germany). The test bars were of a size of 10 mm × 5 mm × 1.5 mm and placed on a tensile geometry configuration. The temperature used in the experiment ranged from −100 °C to 170 °C with a heating rate of 2 °C/min and frequency of 1 Hz.

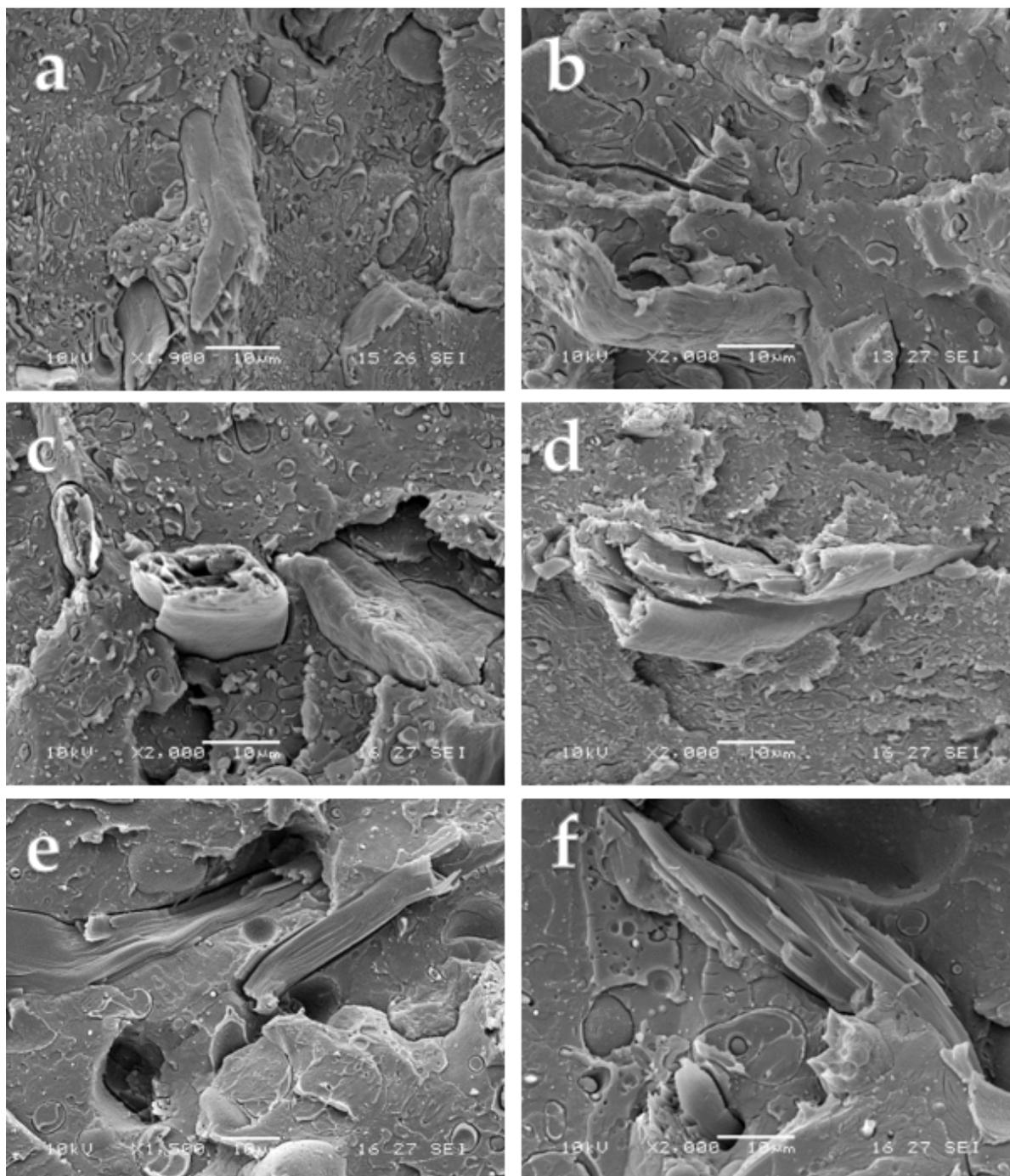
The morphology of the composites was studied by scanning electron microscopy (SEM) using JEOL JSM-5600LV (Tokyo, Japan) and by analyzing the fractured surfaces of the samples obtained by breaking them in liquid nitrogen. Prior to SEM analysis, all the surfaces were sputtered with gold.

## 3. Results

### 3.1. Morphological Evaluation

In previous works [34,35], it was found that a blend with 60 wt. % PLA and 40 wt. % PC was characterized by a co-continuous morphology. This morphological structure was also confirmed using secondary raw materials, as in the current paper.

Figure 2a,c,e refers to the biocomposites produced without the use of the catalytic system, whereas the cryofractured surfaces captured by micrographs in Figure 2b,d,f belong to the biocomposites obtained by reactive extrusion with the compatibilizer.



**Figure 2.** SEM micrographs of cryofractured surfaces of (a) R-PLA60/R-PC40+10ArbI; (b) R-PLA60/R-PC40+10ArbII; (c) R-PLA60/R-PC40+10Ly; (d) R-PLA60/R-PC40+10ArbI+CATA; (e) R-PLA60/R-PC40+10ArbII+CATA; (f) R-PLA60/R-PC40+10Ly+CATA.

A higher adhesion can be achieved in the presence of TA and TBATPB between matrix and cellulose fibers due to the grafting of PLA and PC chains onto cellulose despite the very short reactive extrusion time; indeed, for the micrographs on the left (without CATA) similar to fibers, gaps were also observed between the R-PLA and R-PC. The results obtained are consistent with those in the literature, in which a lack of adhesion was encountered in similar composite materials.

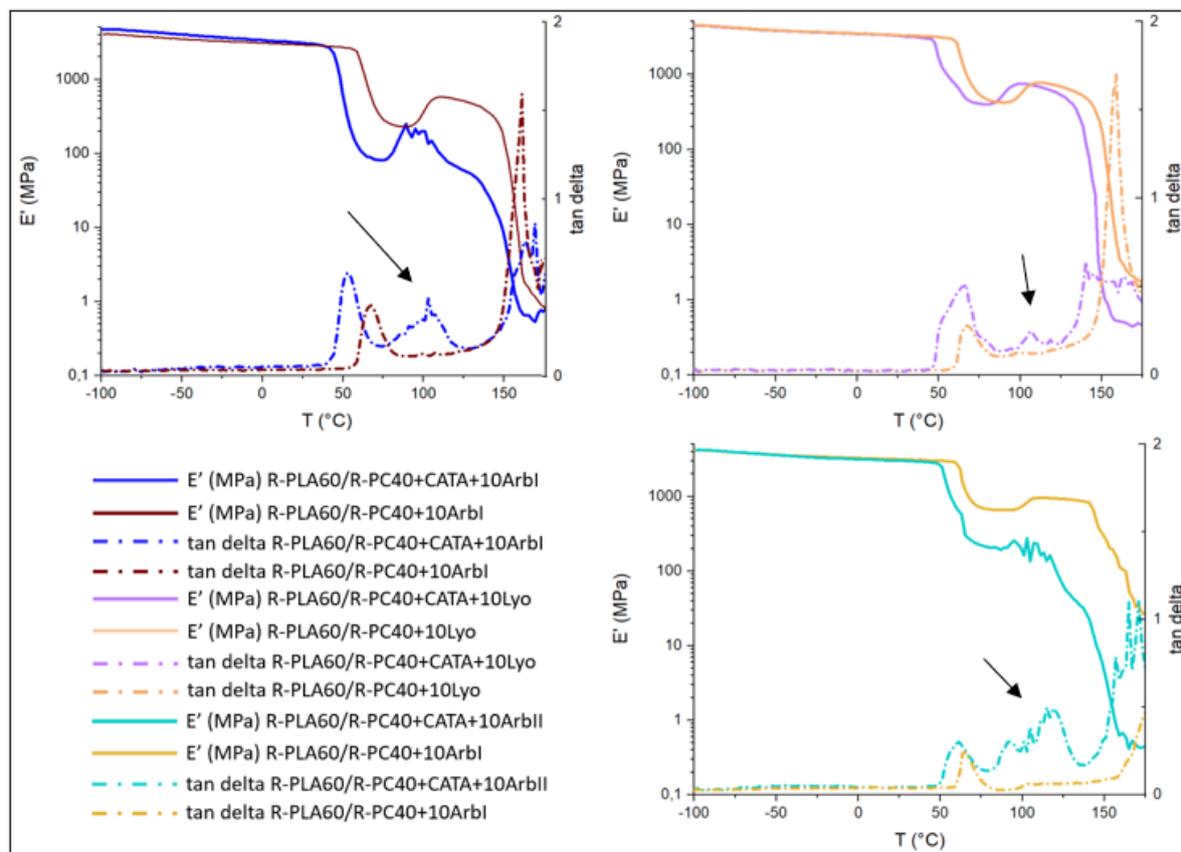
In addition, as reported in a previous study [33], TA, the major component in terms of molecular weight, reacts with the OH groups of polylactic acid and polycarbonate in the

melt phase and, contemporarily, TBATBP forms an active derivative with one, two, or three hydroxyl groups (glycerol). A similar reaction (in the heterogeneous phase) occurs between triacetin and hydroxyl groups on the microcellulose surface, leading to the formation of fiber-bound triacetin. Block copolymers with branches of different lengths can be formed by replacing modified PLA with PC. These PLA-PC copolymer molecules can react with the hydroxyl groups on the cellulose surface in a heterogeneous phase, enabling the grafting of PLA-PC macromolecules onto the cellulose surface.

The study of interfacial adhesion is a well-known problem when natural fibers and synthetic polymers are used [36,37]. In order to obtain a composite with distinctive mechanical properties and good stress transfer from the matrix to the fibers, compatibility is required [38]. Taking into account the peculiarities of each individual fiber type, it can be seen that they all tend to “tangle” when following the well-known phenomenon of the waviness of natural fibers due to the processing operations [15,39], thus providing less reinforcement than would be expected if they had remained intact. In particular, the Lyocell fibers (Figure 2e,f) show better dispersion as they are reworked and deprived of lignin.

### 3.2. DMTA Analysis

The dynamic-mechanical tests reported in Figure 3 show significant results. The most evident is, as seen for pure polymers by Phuong et al. [32], even when using granules from waste, the catalytic system acts to form a copolymer whose  $T_g$  is an intermediate temperature between those of PLA and PC, as evidenced by the tan delta peak represented with the black arrows (more marked for the two types of short Arbocel fibers, less for Lyocell) at about 110 °C.



**Figure 3.**  $E'$  and  $\tan \delta$  evaluation sweeping temperature for all the biocomposites produced. The black arrows represent  $\tan \delta$  peaks.

As for the other  $\tan \delta$  peaks, first, it can be concluded that TA and TBATPB also act as a plasticizer by lowering the glass transition of PLA by about 5 degrees, as found in other systems with plasticized PLA [40,41]. The peaks at about 55 and 60 °C are referred to as PLA glass transition; the peaks around 160° for the biocomposites with CATA and 165 °C for those without CATA are attributable to the PC transition.

Considering the drop-down of elastic modulus as a function of temperature, this occurs at the T<sub>g</sub> of the PLA, but then it rises again due to crystallization [42].

### 3.3. Tensile and Impact Properties

The mechanical tests reported in Figure 4 show that thanks to the introduction of microcellulosic fibers, the stiffness moves above 4 GPa for biocomposites without the use of the compatibilizer, which leads to a decrease in the elastic modulus. Both mechanical strength and stiffness are further improved by the introduction of Arbocel II fibers, which, having an aspect ratio of 10, are probably able to act as reinforcement without an excessive waviness effect, which leads to a decrease as well as the breakage of longer and thinner fibers such as lyocell ones [15]. It should be noted that the catalytic system has an advantageous effect on the impact properties where there is a slight increment, probably due to greater PLA/PC/fiber compatibility. Interesting is the Charpy result of R-PLA60/R-PC40 + CATA + 10LyO, where there is the achievement of 3.3 kJ/m<sup>2</sup>, which is reflected in the stiffness values, which are the lowest of all formulations. Therefore, they ensure greater conservation of energy at the moment of the impact, probably acting as a higher obstacle to crack propagation.

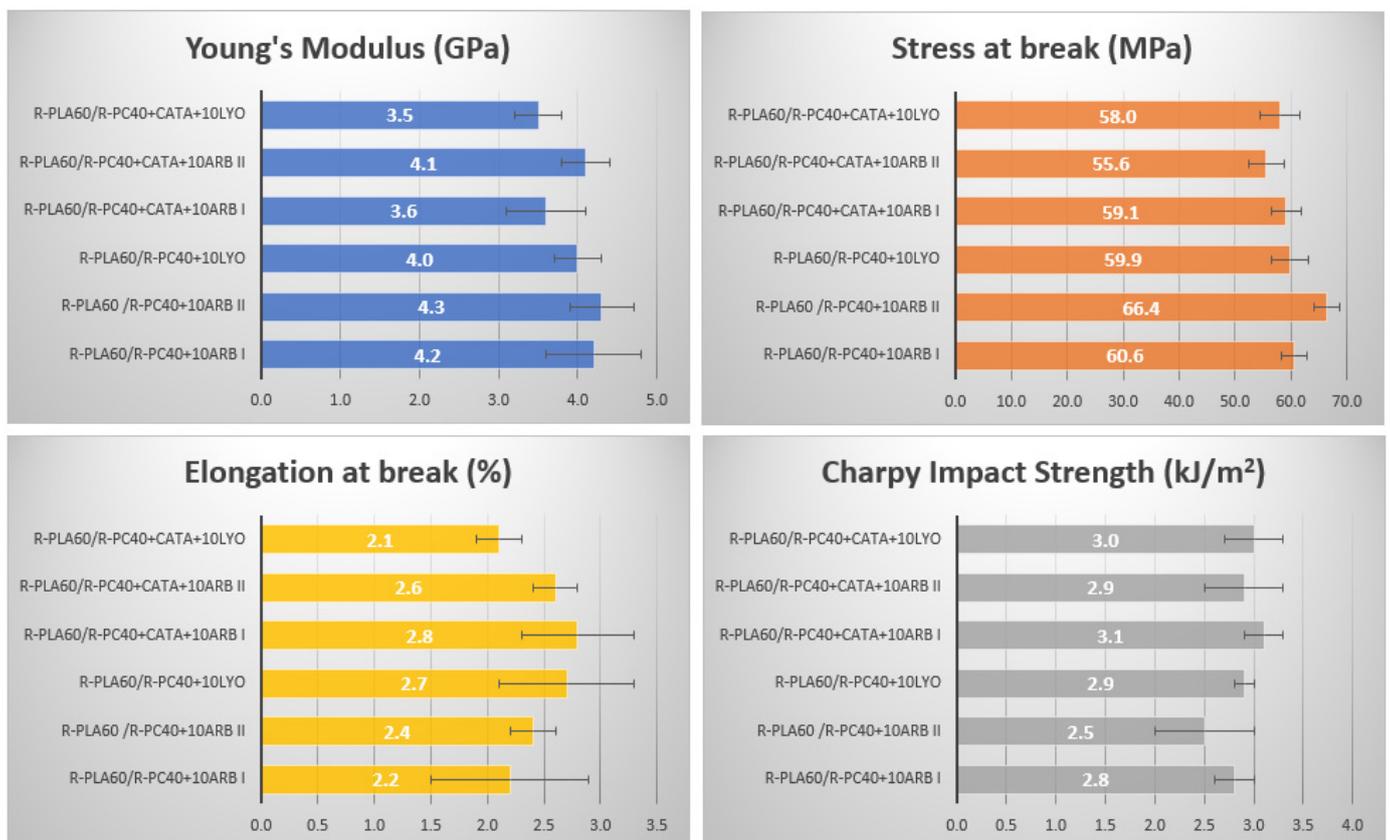


Figure 4. Mechanical properties of R-PLA/R-PC based biocomposites.

### 3.4. Elastic Modulus Predictive Models Comparison

It is not straightforward to evaluate a system based on a thermoplastic matrix in which short-fibers are randomly dispersed to predict the modulus of elasticity. In this case, a large number of geometric, local and mechanical parameters are required [43].

Micromechanical analysis has limitations. A prerequisite for a correct analysis is the assumption that fiber and matrix have perfect adhesion, which is not completely feasible in composites such as those developed in this work [44]. Moreover, the literature data show that the gap between the values calculated on the basis of micromechanical analysis and the experimental data obtained becomes wider when dealing with composites reinforced with natural fibers of plant origin [45].

To overcome these drawbacks, several other prediction methods have been introduced that attempt to consider parameters based on the reinforcement present. These estimations suffer from approximations related, for example, to residual stresses, moisture and voids, but the analytically estimated values, however, can be used for a rough prediction useful for design purposes and to compare quickly and easily different solutions [46]. Referring to the elastic modulus, the models listed in Table 2 consider, in particular, the aspect ratio, the packing factor and Poisson's ratio in order to better predict the elastic modulus of composites containing an increasing amount of reinforcement. In summary, an attempt was made to apply the well-known models of:

- Cox [47]
- Halpin-Tsai [48]
- Clyne [49]
- Starink [50]
- Kim [51]

**Table 2.** List of the analytical expressions used in this work for the prediction of the biocomposites stiffness.

Model	$E_{\text{composite}}$
Cox	$E_c = (1 - V_f)E_m + V_f E_f \cdot \left(1 - \frac{\tanh(na_r)}{(na_r)}\right)$
Halpin-Tsai	$E_c = \frac{3}{8} \left( E_m \cdot \frac{1 + 2 \cdot a_r \cdot \left(\frac{E_f}{E_m} - 1\right) \cdot V_f}{1 - \left(\frac{E_f}{E_m} - 1\right) \cdot V_f} \right) + \frac{5}{8} \left( E_m \cdot \frac{1 + 2 \cdot a_r \cdot \left(\frac{E_f}{E_m} - 1\right) \cdot V_f}{1 - \left(\frac{E_f}{E_m} - 1\right) \cdot V_f} \right)$
Clyne	$E_c = V_f E_f \left\{ \left[ 1 + \left( \frac{0.5 \cdot [E_f(1 - \text{sech}(na_r) + E_m)]}{E_f} \right) - 1 \right] \cdot \frac{\tanh(na_r)}{(na_r)} \right\} + (1 - V_f)E_m$
Starink	$E_c = (1 - V_f)E_m + V_f E_f \cdot \left[ 1 + \left( \frac{E_m - 1}{2 - \text{sech}(na_r)} \right) \frac{\tanh(na_r)}{(na_r)} \right]$
Kim	$E_c = (1 - V_f)E_m + V_f E_f \cdot \left\{ 1 + \left( \sqrt{\frac{E_m}{E_f}} - 1 \right) \cdot \frac{\tanh(na_r)}{(na_r)} \right\}$

For the properties of the fibers, as such, values found in the literature were taken [52,53]. To use these analytical methods, it was necessary to calculate the elastic moduli of the R-PLA/R-PC matrices (with and without CATA) by assessing them to be 3.1 and 3.4 GPa, respectively.

In Table 2,  $E_f$  and  $E_m$  are the elastic moduli of the fibers and matrix, respectively,  $V_f$  is the volume fraction of the fibers and  $a_r$  is the aspect ratio. The dimensional parameter  $n$

is dependent on the Poisson ratio of the matrix (taken as 0.35), and  $p$  is the fibers packing factor having the value of  $2\pi/\sqrt{3}$ .

The Halpin-Tsai model (green bar in Figure 5) is closest to the experimental data in all three biocomposite types, with and without CATA. In fact, in a random-oriented natural short-fiber system like the one considered, the ability of this model to split the elastic modulus into two contributions (longitudinal and transversal) and then provide an empirical equation to monitor the random orientation of the fibers probably provides a more accurate response than the other equations, with respect to a random-oriented natural short-fiber system as demonstrated in the literature [54]. For longer fibers such as Lyocell and Arb II, Cox’s model, which considers discontinuous fibers fully embedded in a continuous matrix assuming that no stress is transferred through the fiber ends, is not so different from the experimental data. On the other hand, the Cox model does not fit very well the composites with Arb I having a very low aspect ratio; the reason for this misfit is strictly correlated to the Cox model assumption that neglects the stress at the fiber’s ends, while it has been demonstrated that for short-fiber composites systems, the fiber stress does not fall to zero [55]. The Clyne and Starink equations take Cox’s model as a starting point, but represent an evolution of it; indeed, they do not want to neglect the stresses at the ends of the fiber (assuming an intermediate value between the average of the load calculated in the matrix away from the interface and the peak it assumes at the interface of the fiber itself). This modification, however, does not guarantee a precise prediction of the modulus of extra short-fiber composites.

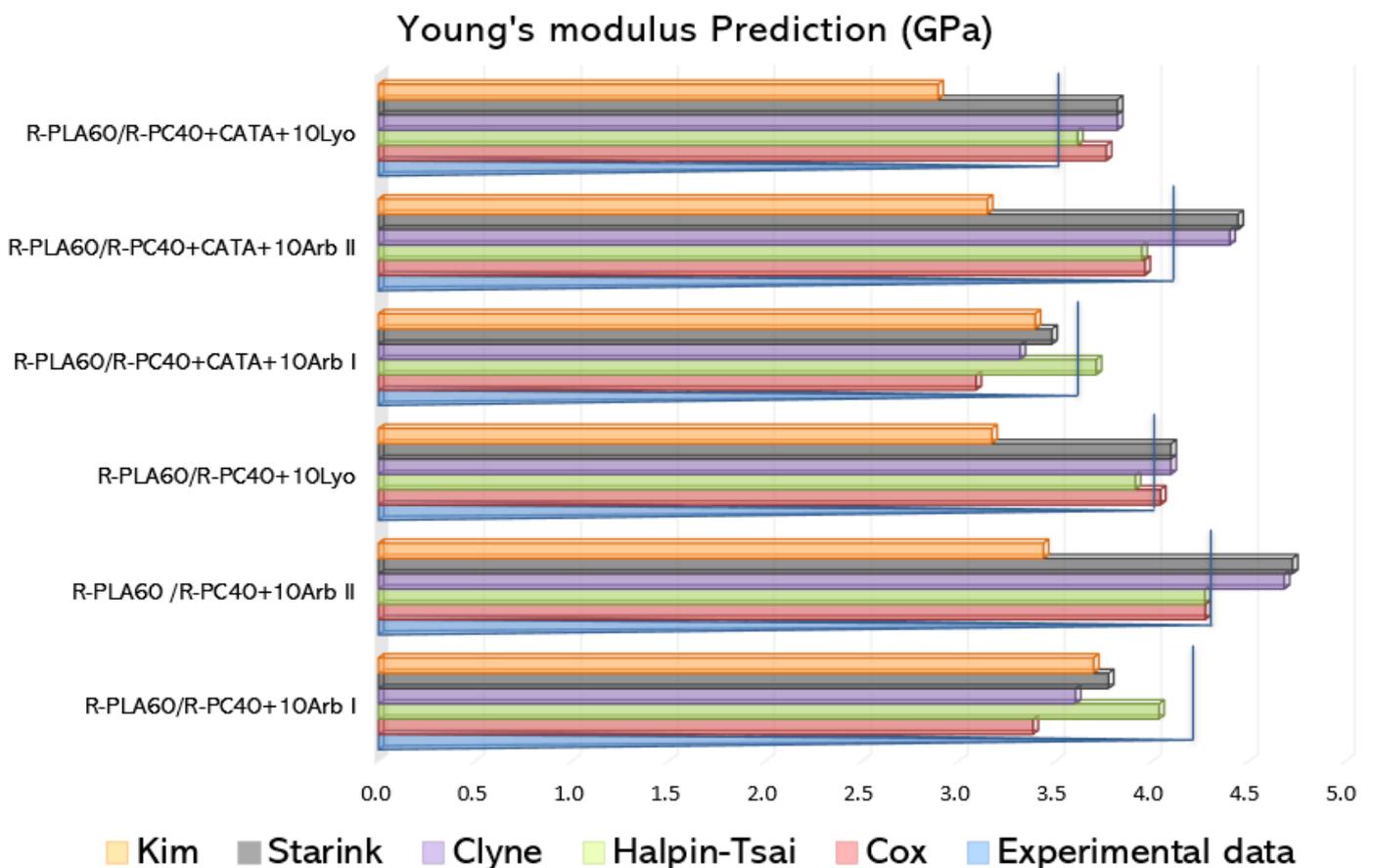


Figure 5. General summary and comparison of predictive models/experimental data.

#### 4. Conclusions

The development of ecological, recyclable, environmentally friendly materials, but at the same time with good mechanical properties, affordable and able to replace petroleum derivatives, is the path that polymer research is currently pursuing. The biocomposites developed in this study, starting from recycled polymers, therefore, represent a valid solution to the problems of post-consumer disposal because they have shown good technical potential related to eco-sustainability.

The recycled polymers, R-PLA and R-PC, do not show qualities that are so dissimilar to virgin polymer blends analyzed in the literature. Therefore, biocomposites from completely recycled processes were made with the addition of cellulosic fibers with three different aspect ratios to investigate how this parameter affected the final properties. The selected matrix, RPLA60/RPC40 containing 60% w/w R-PLA and 40% R-PC, was subjected to reactive extrusion with a catalytic system that favors interchange reactions, allowing the formation of a new species, which has characteristics intermediate between PLA and PC, as demonstrated by DMTA analysis and improving interactions between the matrix and dispersed fibers.

The highest stiffness value was achieved with the Arb II fibers, i.e., those with the longest length (200 microns) and intermediate aspect ratio, showing that not only the length but also the thickness of the natural fiber is a key parameter for increasing the elastic modulus. By inserting the catalyst, these values are lowered, but the Charpy Impact Strength is instead increased due to greater compatibility of the R-PLA/R-PC/fiber system. A study was also carried out concerning the possibility of defining which, among a series of analytical equations, provided the closest fit to the experimental data. It was found that the Halpin-Tsai Model, which “splits” the Elastic Modulus of a biocomposite into two components (Longitudinal and Transverse), was the one that provided values most like those obtained in experimental tests for biocomposites with higher aspect ratios.

**Author Contributions:** Conceptualization, M.-B.C. and A.L.; methodology, V.G. and L.A.; validation, V.G. and L.A.; investigation, V.G. and L.A.; resources, M.-B.C. and A.L.; data curation, V.G. and L.A.; writing—original draft preparation, V.G. and L.A.; writing—review and editing, M.-B.C. and A.L.; supervision, M.-B.C. and A.L.; project administration, A.L.; funding acquisition, A.L. All authors have read and agreed to the published version of the manuscript.

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**Conflicts of Interest:** The authors declare no conflict of interest.

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