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Novel PBAT-Based Biocomposites Reinforced with Bioresorbable Phosphate Glass Microparticles

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Biocomposites based on poly(butylene adipate terephthalate) (PBAT) and reinforced with micro-particles of inorganic biodegradable phosphate glass (PG) at 2, 10, and 40 wt% are prepared and characterized from a mechanical and morphological point of view. Scanning electron microscope (SEM) images show a good dispersion of the PG micro-grains, even at high concentrations, in the PBAT matrix, resulting in homogeneous composites. Tensile and dynamic-mechanical tests, respectively, indicate that Young's and storage moduli increase with PG concentration. The reinforcement of PBAT aims at modifying and tailoring the mechanical and viscoelastic properties of the material to expand its application field especially in the food and agricultural packaging sector, thanks to the similarity of PBAT performance with polyethylene.

1. Introduction

In the last century, plastic materials have been rapidly developed and widely employed in varieties of fields thanks to their peculiar properties and low costs. Unfortunately, most of these conventional plastics, such as polyethylene (PE), polypropylene (PP), and polystyrene (PS) have oil origin and their wastes cannot be biodegraded. Therefore, the increase in the production and consumption of traditional plastics results in oil consumption and serious environmental pollution. These major problems caused by conventional plastics should be solved for sustainable development in future.[1]

The present work aims at the preparation and subsequent microstructural, mechanical, and dynamic-mechanical characterization of biocomposites based on poly(butylene adipate terephthalate) (PBAT), loaded with micro-particles of inorganic biodegradable phosphate glass (PG)[2] at 0, 2, 10, and 40 wt%, respectively named as PBAT, PBAT+2%PG, PBAT+10%PG, and PBAT+40%PG.

The reinforcement of flexible polymeric matrices such as PBAT has the purpose of modifying and tailoring the mechanical and viscoelastic properties of the material in order to expand its application field especially in the food and agricultural packaging sector,[3] thanks to the similarity of PBAT performance with PE.[4]

PBAT is an oil-based polymer that is expected to undergo complete biodegradation in soil over a period of 6 weeks.[5] This behavior makes it an ideal sustainable replacement for PE. Moreover, experimental evidence shows the possibility to obtain PBAT monomers from renewable sources, making this polymer even more interesting in terms of environmental sustainability.[6,7,8]

The PBAT-PG master batches are prepared by solvent casting, using chloroform as suitable solvent. Following a thermal drying process up to constant weight, the resulting composite pellets are used for the injection molding of model 1BA specimens, employed for tensile characterization, according to UNI EN ISO 527 standard.

The specimens are subjected to uniaxial tensile stress and the collected data are analyzed to obtain characteristic parameters such as the Young's modulus (E), the yield stress (σ_Y), the stress at break (σ_B), the elongation at break (ϵ_B), and the toughness (T).

Following the ASTM D7028 standard, dynamic-mechanical analysis (DMA) is carried out on a DMA analyzer using a single cantilever clamp to determine the storage modulus (E'), loss modulus (E''), and loss factor (tanδ) of the composites.

The sample microstructure is then observed on cross-sectional surface by means of SEM.

2. Results and Discussion

SEM images display homogeneous dispersion and distribution of the filler particles in the polymer matrix with no aggregates or

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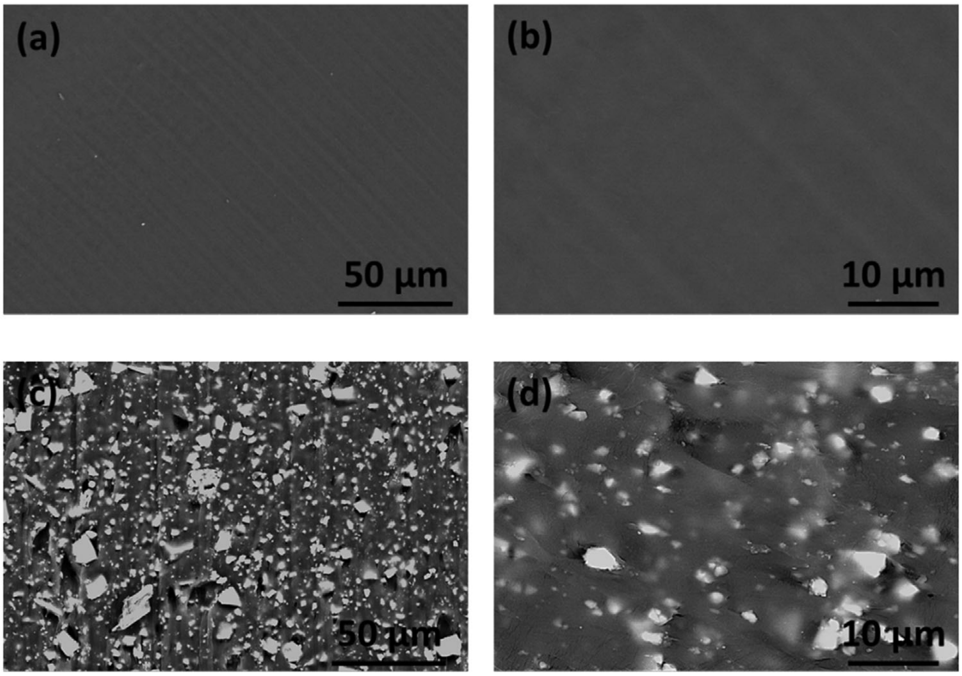

Figure 1. SEM images of PBAT (a, b) and PBAT + 40% PG (c, d) at different magnifications, as representative samples.

Table 1. Tensile properties of PBAT-PG composites.

Sample	E [MPa]	σ_Y [MPa]	σ_B [MPa]	ϵ_B [mm mm^{-1}]	T [MJ m^{-3}]
PBAT	102 ± 20	8.9 ± 0.5	13.9 ± 0.9	3.7 ± 0.5	41 ± 7
PBAT+2%PG	104 ± 15	9.0 ± 0.2	13.8 ± 0.2	3.9 ± 0.2	41 ± 2
PBAT+10%PG	123 ± 15	8.4 ± 0.2	12.7 ± 0.4	3.5 ± 0.4	35 ± 2
PBAT+40%PG	186 ± 30	7.2 ± 0.2	7.6 ± 0.7	1.7 ± 0.2	12 ± 3

phase separation that would cause a deterioration of the material properties (**Figure 1**).

PG particles show irregular geometry and are generally smaller than 10 µm in size. As the magnification increases, no voids at the interface can be noted, indicating a good surface wettability and compatibility of PG particles by the polymer matrix.

PBAT-PG composites exhibit an effective increase of E up to 82% with increasing the PG content, while showing a reduction of σ_Y up to 20%, of σ_B up to 46%, of ϵ_B up to 57%, and of T up to 72%, as summarized in **Table 1**.

The increase of E and the limited decrease in σ_Y with increasing the PG content suggest the activation of a stress transfer mechanism across the PBAT-PG interface, confirming a slight positive interaction between PBAT and PG.[9]

ϵ_B and T continuously decrease as the amount of PG in the polymer matrix increases, since PG particles are stiff and no deformable, according to the general trend observed for filler characteristics on the polymer properties.[10,11]

DMA results as a function of temperature for the different composites are shown in **Table 2**. E' increases with increasing PG content within the whole analyzed temperature range. In particular, above the glass transition temperature (T_g), E' enhancement can be ascribed to the decrease in the segmental mobility of the polymer chains, due to the presence of PG micro-particles. Due to the higher stiffness of PG, PBAT-based composites have greater resistance to macromolecular motions under stress conditions if compared with unfilled polymer.[12]

T_g is identified as the temperature corresponding to the peak of the tanδ curve, obtained by the E''/E' ratio. The PG addition does not significantly change the T_g value of pristine PBAT.

A very similar trend was observed in other biocomposite systems reinforced with natural fibers, suggesting a restricted interaction between filler and polymer matrix, as already observed.[13,14]

3. Conclusion

PBAT is considered as one of the most promising biodegradable polyesters and this work demonstrates the successful realization of biodegradable PBAT-based composite materials reinforced by PG micro-particles.

According to SEM images of the specimens, the filler particles are finely dispersed and homogeneously distributed within the polymer matrix, even at the highest concentration, without agglomerates formation.

The results of the tensile tests reveal an effective increase in the stiffness of the composites compared to the pure polymer, at the expense of strength and elongation at break and toughness.

DMA characteristics of the composites see an increase in the storage modulus as the PG concentration increases, while the glass transition temperature remains substantially constant.

The developed materials prove to be valid biodegradable and eco-friendly alternatives to traditional thermoplastic polymers, such as PE, and can be applied in many fields, especially in package and mulch film applications.

Table 2. Dynamic-mechanical properties of PBAT-PG composites.

Sample	E' (-40°C) [MPa]	*E'* (10°C) [MPa]	*E'* (20°C) [MPa]	*E'* (40°C) [MPa]	T_g [°C]
PBAT	2800 ± 20	203 ± 20	178 ± 20	135 ± 20	−17.8 ± 0.5
PBAT+2%PG	2995 ± 20	251 ± 20	223 ± 20	181 ± 20	−17.4 ± 0.5
PBAT+10%PG	3310 ± 20	284 ± 20	252 ± 20	206 ± 20	−16.9 ± 0.5
PBAT+40%PG	5350 ± 20	575 ± 20	496 ± 20	359 ± 20	−17.2 ± 0.5

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

biopolymers, composites, phosphate glass, poly(butylene adipate terephthalate)

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