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Novel PBAT-Based Biocomposites Reinforced with Bioreposable Phosphate Glass Microparticles

Elena Togliatti, Diego Pugliese, Alberto Giubilini, Massimo Messori, Daniel Milanese, and Corrado Sciancalepore*

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, thanks to the similarity of PBAT performance with polyethylene. 

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.

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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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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 $\sigma_y$ up to 20%, of $\sigma_B$ up to 46%, of $\epsilon_B$ up to 57%, and of $T$ up to 72%, as summarized in Table 1.

The increase of $E$ and the limited decrease in $\sigma_y$ 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.$^{[9]}$

$\epsilon_B$ 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 packaging and mulch film applications.
Table 2. Dynamic-mechanical properties of PBAT-PG composites.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$E'$ (-40°C) [MPa]</th>
<th>$E'$ (10°C) [MPa]</th>
<th>$E'$ (20°C) [MPa]</th>
<th>$E'$ (40°C) [MPa]</th>
<th>$T_g$ [°C]</th>
</tr>
</thead>
<tbody>
<tr>
<td>PBAT</td>
<td>2800 ± 20</td>
<td>203 ± 20</td>
<td>178 ± 20</td>
<td>135 ± 20</td>
<td>−17.8 ± 0.5</td>
</tr>
<tr>
<td>PBAT+2%PG</td>
<td>2995 ± 20</td>
<td>251 ± 20</td>
<td>223 ± 20</td>
<td>181 ± 20</td>
<td>−17.4 ± 0.5</td>
</tr>
<tr>
<td>PBAT+10%PG</td>
<td>3310 ± 20</td>
<td>284 ± 20</td>
<td>252 ± 20</td>
<td>206 ± 20</td>
<td>−16.9 ± 0.5</td>
</tr>
<tr>
<td>PBAT+40%PG</td>
<td>5350 ± 20</td>
<td>575 ± 20</td>
<td>496 ± 20</td>
<td>359 ± 20</td>
<td>−17.2 ± 0.5</td>
</tr>
</tbody>
</table>

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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