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Filler Content Effect on Water Uptake and Thermal Stability of Poly(3-Hydroxybutyrate-Co-3-Hydroxyhexanoate)/Microcrystalline Cellulose Biocomposites

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In this study, biocomposites of poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) (PHBHHx)/cellulose microcrystalline (MCC) extracted from olive husk flour are prepared by melt compounding at various filler content ratios, i.e., 10, 20, and 30 wt%. The effect of the MCC content on the morphology, thermal stability, crystallinity, and water uptake of the PHBHHx biocomposites is investigated. The results showed that the addition of MCC to PHBHHx decreased the thermal stability of the biocomposites compared to that of neat polymer, however more pronounced at a higher filler content. This is due probably to the tendency of MCC particles to agglomerate, inducing heterogeneities and defects within the polymer matrix observed by scanning electron microscopy. Furthermore, both crystallinity and water uptake after 24 h of immersion increased with the filler content.

1. Introduction

The interest in using sustainable materials, coming from renewable resources and/or biodegradable, is growing rapidly due to the ecological context.[1–4] In this regard, polyhydroxyalkanoates (PHAs), generated by microorganisms, have highly attracted attention in various fields due to their physico-mechanical properties, which are comparable to those of polyolefins or PET.[5,6] However, the application of PHAs to some industrial fields is rather limited due mainly to its slow crystallization rate, high crystallinity, and high thermal sensitivity.[7,8] Among the family of PHAs, PHBHHx copolymer is considered as the third generation one; it is the most potential biopolymers to substitute traditional commodity plastics. It is fully biodegradable and biocompatible. Due to its ductile nature and wider processing window compared to poly(3-hydroxybutyrate), PHBHHx has a broad range of applications in various industries such as biomedical sector including tissue engineering, bio-implant patches, drug delivery, surgery, wound dressing and scaffolds, agriculture in the controlled release of insecticides and also flexible packaging.[8] Together with the development of biocomposite materials, the addition of cellulosic fillers to PHAs could be one of the most promising routes to further improve their properties.[9] Indeed, cellulose due to its abundant availability, renewability, biodegradability, high strength, and stiffness could replace advantageously layered silicates, carbon nanomaterials, and other metallic oxide fillers.[10] Natural fiber-reinforced biocomposites have been used for many applications and accepted by consumers, especially in the automotive industry (interior products such as door panels, seat backs, headliners, package trays, dashboards, and interior parts), packaging and building industries, where a high load carrying capacity is not required.[11,12] Therefore, this work aims to investigate the effect of MCC content at 10, 20, and 30 wt% on the morphology, thermal stability, crystallinity, and water uptake of PHBHHx biocomposites prepared by melt compounding. The properties performances of the PHBHHx/MCC biocomposites will be discussed based on the neat polymer.

2. Experimental Section

2.1. Materials Used

PHBHHx containing 11% of hydroxyhexanoate in granular form was manufactured by Kaneka Corporation (Osaka, Japan) under the grade Aonilex X151A. According to the manufacturer, the polymer has a density = 1.19 g cm^{-3}, a melting temperature (T_m) = 130 °C, a glass transition temperature (T_g) = −0.2 °C, and a crystallinity index (X_c) = 34%. MCC was extracted from olive husk flour, which has been collected in the region of Bejaia in Algeria. The detailed extraction procedure of MCC is reported elsewhere.[13] The size fraction, selected after sifting, has a maximum average diameter of 50 µm. This is more appropriate to have a good dispersion of the filler within the thermoplastic matrix.[14] The particle shape of MCC is generally rod-like having different sizes ranging from a few microns to 50 µm as shown in **Figure 1**.

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