Composite Interfaces, Vol. , No. , pp. 1–19 (2007)

Also available online - www.brill.nl/ci

Cellulose-based biocomposites: comparison of different multiphasic systems

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Received 5 September 2006; accepted 1 February 2007

Abstract—Biocomposites (biodegradable composites) are obtained by blending biodegradable polymers and fillers. Since the main components are biodegradable, the composite as a whole is also expected to be biodegradable. This paper presents various biocomposites that have been elaborated with cellulose or lignocellulose fibers from diverse sources, with different lignin contents. This paper is targeted on the analysis of 'fiber–matrix' interactions of two types of biocomposites based on agropolymer (plasticized wheat starch) and biopolyester (polybutylene adipate-*co*-terephthalate), named APB and BPB, respectively. Processing and main properties of both biocomposites are shown and compared. Polyolefin-based composite (PPC), which is known to present very poor 'fiber–matrix' interactions, is used as a reference. Through the Young's modulus, mechanical properties have shown that the reinforcement, by increasing fiber content, is much more significant for APB compared to BPB. The evolution of chains mobility, evidenced through shift of T_g values, according to the increase in fiber content and thence in interfacial area, have shown that the fiber–matrix interactions are higher for APB. BPB presents intermediate values, higher than PPC ones. These results are in agreement with the analysis of the composite morphologies performed by SEM on cryogenic fractures. Finally, by determining the theoretical works of adhesion and the interfacial tensions from contact angle measurements, it is shown that these parameters are partially able to predict the level of interaction between the fibers and the matrix. We could show that the perspectives of such work seem to be of importance to tailor new materials with a controlled end-use.

Keywords: Biocomposites; plasticized starch; biopolyester; cellulose; lignocellulose; fiber; multiphase system; interaction.

1. INTRODUCTION

Biocomposites (biodegradable composites) are a special class of composite materials. They are obtained by blending biodegradable polymers with biodegradable fillers (e.g. lignocellulose fibers). Since the main components are biodegradable, the

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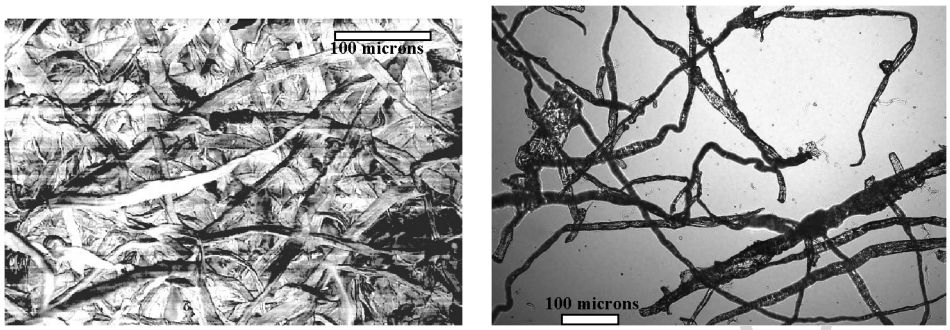
composite as a whole is also expected to be biodegradable. Tailoring new composites within a perspective of eco-design or sustainable development is a philosophy that is applied to more and more materials. Ecological concerns have resulted in a resumed interest in renewable resources-based and/or compostable products. This is the reason why material components such as natural fibers and other biodegradable polymers can be considered as 'interesting' – environmentally safe – alternatives.

A classification of these polymers has been shown elsewhere [1]. The main groups are (i) the agro-polymers (polysaccharides, proteins) and (ii) the biopolyesters (biodegradable polyesters) such as PLA (polylactic acid), PHA (polyhydroxyalkanoate) or aromatic and aliphatic copolyesters. Biodegradable polymers show a large range of properties and, at present, they can compete with non-biodegradable thermoplastics in different fields (e.g. packaging, agriculture, hygiene or cutlery).

Cellulose or lignocellulose fibers are widely used as biodegradable fillers. With their environmentally friendly character and some strong techno-economical advantages, these fibers motivate more and more different industrial sectors (e.g. automotive) to replace, for example, common fiberglass. Intrinsically, the agro-fibers have a number of interesting mechanical and physical properties [2–6]. The structure and the organization of such agro-materials have been extensively described in different publications [2, 4]. But till now, the macromolecular architectures in the fiber cell walls still remain partially unknown. The main macromolecular elements are cellulose, hemicellulose and lignin. According to botanical sources, these renewable fillers present large variations in their composition [7]. The different contents can also vary according to the conditions of the fibers' pre-treatment (e.g. fractionation process, surface treatment) [8, 9].

For short-term applications with a controlled end of life, biocomposites present strong advantages. Thus, a large number of papers have been published on this topic. A few publications are based on agropolymer matrices, such as plasticized starch [10–12] or proteins [13], and on chemically modified agropolymers (e.g. cellulose acetate) [15–17]. However, most of the studies are focused on biopolyester matrices [1, 18, 19], which are numerous and largely available. A great number of biopolyesters have been investigated in association with diverse lignocellulose fillers, from various origins. The main matrices are PHA [20–39] and PLA [26, 31–34]. Regarding biopolyesters from petroleum resources, aliphatic copolyesters have been associated with bleached cellulose fibers [21], bamboo fibers [35] or flax, oil palm, jute or ramie fibers [21]. Recently, an aromatic copolyester has been associated with lignocellulose fillers from wheat straw [8, 9].

The aim of this paper is focused on the comparison of the behaviors of two different types of biocomposites, agropolymers-based biocomposites (APB) and biopolyesters-based biocomposites (BPB). To estimate the compatibility and interactions between cellulose or lignocellulose fibers and each type of matrix, no compatibilizers have been added. The composites are obtained after different processing steps, such as extrusion and injection molding. The main properties are examined and compared. The filler–matrix interactions are analyzed for both types of multi-



(a) (b)

Figure 1. Cellulose fibers: SEM (a) and optic (b) micrographs.

phase systems (APB and BPB) in connection with the filler and the matrix nature. Besides, we have compared the biocomposites with composites based on a PP matrix (PPC) which, as is well known, show very poor matrix–fiber compatibility in the absence of compatibilizer. PPC is only used as a reference material. Three main approaches are used to investigate the 'fiber–matrix' interactions; (i) the mechanical properties through the evolution of the modulus with the fibers content, (ii) the glass temperature shift with the increase of filler content and then (iii) the morphology of the composites.

2. MATERIALS AND PROCESS

2.1. Different fibers

Two types of fibers are tested, cellulose and lignocellulose fibers. The density of these fibers is between 1.50 (cellulose) and 1.45 (lignocellulose).

2.1.1. Cellulose-based fibers. Cellulose fibers (CF) from leafwood are supplied by Omya Rettenmier (JRS-Arbocel, Germany). Different semi-crystalline cellulose microfibers with increasing lengths are tested. Figure 1(a,b) shows on SEM and optic microscopy micrographs (Fig. 1(a) and 1(b), respectively) that the fibers are like ribbons. Initial average lengths are respectively 60 (sCF), 300 (mCF) and 900 (lCF) μm. Cellulose content is greater than 99.5%.

2.1.2. Lignocellulose-based fibers. The first type of lignocellulose fibers is paper pulp fibers (PF) supplied by La Rochette (France). PF are obtained from broad–leaved species through a semi-chemical pulping treatment involving a chemical fractionation at 175°C using ammonium sulfite followed by a mechanical treatment in a refiner. Paper pulp is taken from a production line before mixing with recycled fibers and sheet forming. Moisture content is then 65 wt%. To prepare the filler (PF), the pulp sample is rinsed with distilled water and then pressed until residual