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New biocomposites based on thermoplastic starch and bacterial cellulose

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ABSTRACT

Bacterial cellulose, produced by *Acetobacter Xylinum*, was used as reinforcement in composite materials with a starch thermoplastic matrix. The composites were prepared in a single step with cornstarch by adding glycerol/water as the plasticizer and bacterial cellulose (1% and 5% w/w) as the reinforcing agent. Vegetable cellulose was also tested as reinforcement for comparison purposes. These materials were characterized by different techniques, namely TGA, XRD, DMA, tensile tests, SEM and water sorption assays. All composites showed good dispersion of the fibers and a strong adhesion between the fibers and the matrix. The composites prepared with bacterial cellulose displayed better mechanical properties than those with vegetable cellulose fibers. The Young modulus increased by 30 and 17 fold (with 5% fibers), while the elongation at break was reduced from 144% to 24% and 48% with increasing fiber content, respectively for composites with bacterial and vegetable cellulose.

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1. Introduction

New functional materials obtained from renewable resources have gained much attention in the last decades due to the global increasing demand for alternatives to fossil resources [1]. For example, composites based on thermoplastic polymers and natural fibers are very attractive materials because of their good mechanical properties, sustainability and environmental-friendly connotation [2]. These materials have been widely used in the automotive and packaging industries [2] and their applications in other areas are being actively sought.

The use of natural fibers as reinforcing elements in composite materials presents important advantages, when compared with their synthetic or inorganic counterparts, namely biodegradability, high availability, low cost, low energy consumption, low density, high specific strength and modulus (with fibers possessing an adequate aspect ratio), high sound attenuation and comparatively easy processing ability due to their flexibility and non-abrasive nature [3].

In addition, if the natural fibers are combined with biodegradable matrices, the obtained composite materials are expected to be fully biodegradable. The development of biocomposites gained particular relevance with the increasing availability and diversity of biodegradable polymers [4].

Starch is one of the most abundant natural polymers and is considered as a promising raw material for the development of novel materials, including biocomposites. It can be converted into a thermoplastic material, known as thermoplastic starch (TPS), through the disruption of the molecular chain interactions under specific conditions, in the presence of a plasticizer. Water and glycerol are the most widely used plasticizers in the production of TPS. When natural fibers are mixed with TPS, its mechanical properties are improved, indicating a good adhesion between the reinforcing fibers and the matrix [4,5]. Several studies have been published dealing with the preparation and characterization of TPS-based composites with different cellulose substrates, namely commercial regenerated cellulose fibers [6], vegetable fibers [6–8], microcrystalline cellulose [9], microfibrillated cellulose [3] and cellulose nanocrystallites [5]. Apart from the enhanced mechanical properties of these reinforced TPS materials, a significant improvement in water resistance is also obtained by adding cellulose crystallites [10] or microfibrillated cellulose [3]. In general, these TPS/fiber composites also displayed improved thermal stability due to the higher thermal resistance of cellulose fibers [7].

Owing to its unique properties, such as high mechanical strength, high crystallinity and a highly pure nanofibrillar network structure, bacterial cellulose, produced by *Acetobacter Xylinum*, is becoming a promising biopolymer for several applications. For example, due to its biocompatibility [11], bacterial cellulose has been extensively investigated in the biomedical field [12–14]. Other applications include its use as component for audio membranes [15], electronic paper [16], optically transparent composites

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[17], reinforcing agent for paper [18] and other polymeric materials and composites [19], among others.

However, to the best of our knowledge, only two studies dealing with the preparation and characterization of bacterial cellulose-starch composites have been published so far [20,21]. Grande et al. obtained a plasticized starch/bacterial cellulose nanocomposite sheets by hot-pressing. The ensuing films were characterized in terms of their morphology, whereas other important parameters, such as their mechanical and thermal properties, were not investigated. The incorporation of bacterial cellulose microfibrils, obtained by the acid hydrolysis of the cellulose network, into extruded TPS and starch-pectin blends was also studied [21]. However, in this work, the peculiarity of this cellulose substrate was not fully exploited, since the nano- and micro-fibril three-dimensional network morphology was partially destroyed during the hydrolysis step. Indeed, the microfibrils derived from bacterial cellulose did not improve the modulus to the same extent as cotton or softwood counterparts.

The present paper describes the preparation and characterization of biocomposite materials obtained by the incorporation of bacterial cellulose into a TPS matrix during the gelatinization process. Similar composite materials reinforced with vegetable cellulose fibers were also prepared for comparison purposes.

2. Experimental

2.1. Materials

Bacterial cellulose (BC) (a tridimensional network of nano and microfibrils with 10–100 nm width), in the shredded wet form (95% of humidity), was supplied by Forschungszentrum für

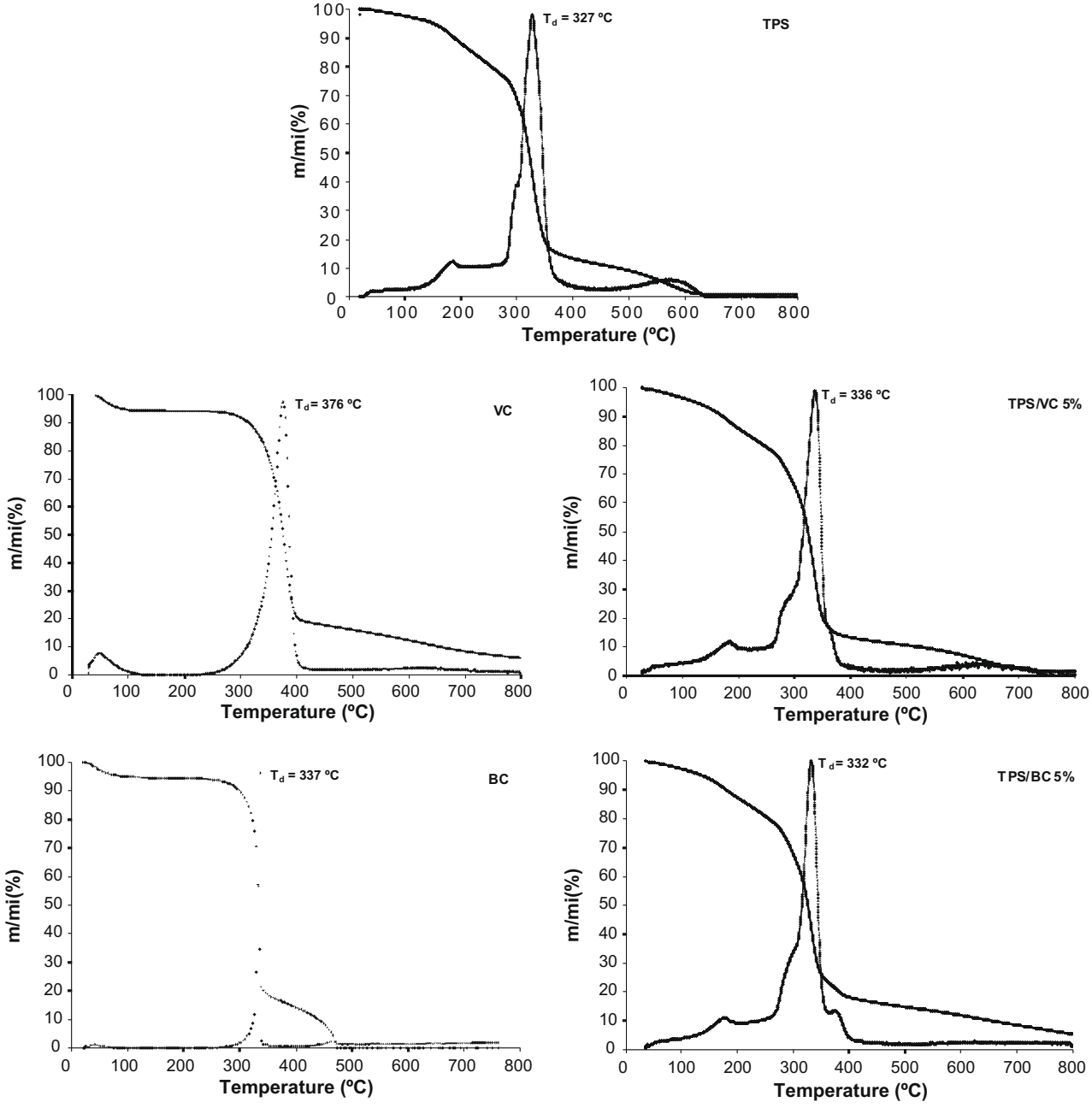

Fig. 1. Thermogravimetric curves for TPS, BC, VC, TPS/BC 5% and TPS/VC 5% composites.

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