

# Plasticized starch–cellulose interactions in polysaccharide composites

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Received 13 November 2000; received in revised form 25 January 2001; accepted 6 February 2001

## Abstract

This paper is focused on the interactions between leafwood cellulose fibres and a plasticized wheat starch matrix. Different plasticized starch (TPS)-based composites have been elaborated. LDPE-based composites are used as reference materials (no fibre–matrix interactions). After extrusion and injection moulding, the properties of the different composites are analysed. Mechanical properties (tensile tests), thermo-mechanical properties (DMTA) and morphology (SEM) are evaluated. DMTA analysis shows for TPS composites a strong evolution of the main relaxation temperature, which can be linked to the existence of cellulose–starch interactions resulting in a decrease of starch chain mobility. This phenomenon is consistent with the evolution of mechanical behaviour. SEM observations correlate this hypothesis. After cryogenic fracture, TPS composites present fibres, which are embedded in the matrix. On the composites, reinforcing effects have been observed according to the evolution of fibre length and fibre content. © 2001 Elsevier Science Ltd. All rights reserved.

**Keywords:** Composite; Thermoplastic starch; Cellulose fibre

## 1. Introduction

In recent years, great progress was achieved in the development of biodegradable products on the basis of agricultural materials [1,2]. Different approaches have been made to use starch for the production of tailored materials [3]. Native starch is transformed to obtain easy processed starch [4–6]. The so-called ‘thermoplastic starch’ (TPS) or plasticized starch is obtained after disruption and plasticization of native starch, with water and plasticizer. Unfortunately, properties of such a product do not fulfil all requirements in some applications such as packaging. To improve the properties, research laboratories have developed two strategies: chain modifications (e.g. acetylation) [7], and starch-based multiphased products. Compostable multilayers [8,9] or blends [10–14] have been developed by different associations between TPS and biodegradable polymers which are mainly biodegradable polyesters. These blends present quite good water resistance but mechanical properties are rather poor. TPS–polyester compatibility is more or less weak [12].

To preserve renewability (renewable raw materials), biodegradability and to improve the mechanical resistance

of the final products, associations between cellulose fibres and TPS have to be tested. Several studies [15–22] and applications (e.g. automotive market) have demonstrated the interest of using cellulose fibres as reinforcement in thermoplastic matrixes but only few papers are focused on polysaccharide-based composites. Some authors [16–22] have shown that cellulose fibres or microfibrils in a TPS matrix improve the tensile strength. According to Frunke et al. (1998) [22], a significant improvement of water resistance is achieved by adding small amounts of commercial cellulose fibres (till 15%). Also, Dufresne et al. (1998, 2000) [19,20] show an improvement of water resistance by reinforcing plasticized starch films with cellulose microfibrils. This behaviour is related to the hydrophobic character of the cellulose fibres in comparison to starch hydrophilic property. Besides, these authors [19] show an improved thermal stability due to a higher and longer rubbery plateau.

The aim of this work is to test the addition of cellulose fibres in a TPS matrix and to report the subsequent properties. This paper is more particularly focused on the interaction between the fibres and the matrix. Various biodegradable composites have been elaborated with a soft TPS matrix and different fibre lengths and contents. Besides, we have used LDPE-based composites as reference materials, where the fibre–matrix interactions may be considered as poor since no specific compatibilizer are added [15–18]. LDPE grade has been chosen to show at

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