Revealing the 3D internal structure of natural polymer microcomposites using X-ray ultra microtomography

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Summary

Properties of composite materials are directly affected by the spatial arrangement of reinforcement and matrix. In this research, partially hydrolysed cellulose microcrystals were used to fabricate polycaprolactone microcomposites. The spatial distribution of cellulose microcrystals was characterized by a newly developed technique of X-ray ultra microscopy and microtomography. The phase and absorption contrast imaging of X-ray ultra microscopy revealed two-dimensional and three-dimensional information on CMC distribution in polymer matrices. The highest contrast and flux (signal-to-noise ratio) were obtained using vanadium foil targets with the accelerating voltage of 30 keV and beam current of >200 nA. The spatial distribution of cellulose microcrystals was correlated to the mechanical properties of the microcomposites. It was observed that heterogeneous distribution and clustering of cellulose microcrystals resulted in degradation of tensile strength and elastic modulus of composites. The utilization of X-ray ultra microscopy can open up new opportunities for composite researchers to explore the internal structure of microcomposites. X-ray ultra microscopy sample preparation is relatively simple in comparison to transmission electron microscopy and the spatial information is gathered at much larger scale.

Introduction

Among various natural reinforcements, much interest has been devoted to the usage of polysaccharides such as chitin, starch and cellulose. Cellulose is the most abundant naturally occurring polymer on earth, and is produced and recycled at a rate of 10^{10} tons year^{-1} (Hon, 1994). It is most widely found in plants (Fleming *et al.,* 2000; Habibi *et al.,* 2008; Siqueira *et al.,* 2009), but some animals (Angles & Dufresne, 2000) and bacteria (Grunert & Winter, 2002) are also sources of cellulose.

Cellulose consists of both crystalline and amorphous domains. Although the amorphous part enables deformation, the crystalline part provides the strength, density and rigidity (Hamad, 2006). The reinforcing efficiency of cellulose depends on the fraction of crystalline domains and their interaction with the domain that they are reinforcing (Lapa *et al.*, 2007). Native cellulose subjected to strong acid hydrolysis readily breaks down into micro- or nanocrystals (Batista, 1975).

Despite the above-mentioned attractive properties, the lack of control over the dispersion of cellulose crystals in polymer matrices has limited their use in the composite industry (Oksman *et al.,* 2006). Cellulose is a hygroscopic linear polysaccharide of β-D-glucopyranose units, which are connected by $(1{\rightarrow}4)$-glycosidic bonds. The strong intermolecular hydrogen bonding forces, arising from the hydroxyl groups on the cellulose chains, cause a major fabrication difficulty. The hydrophilic nature of cellulose and the nonpolar characteristics of most thermoplastics make it difficult to achieve a sufficient dispersion of this reinforcement in polymer matrices.

Conventional methods for characterization of microstructures in composite materials are generally limited to study of the surface or a localized portion of the composite volume. In addition, they often require tedious sample preparation steps, or they are complex in the sense of data interpretation. These methods include: optical microscopy (Wang & Sain, 2007), scanning electron microscopy (SEM) (Choi & Simonsen, 2006), transmission electron microscopy (Bodenson & Oksman, 2007), atomic force microscopy (Matsumura & Glasser, 2000), small angle X-ray scattering (Mele *et al.,* 2002) and wide angle X-ray diffraction (Vaia & Liu, 2002).

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