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Novel automated method for evaluating the morphological changes of cellulose fibres during extrusion-compounding of plastic–matrix composites

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ABSTRACT

A novel method based on fluorescence optical microscopy has been developed for determining the fibre geometrical changes occurring during the melt processing of cellulose-reinforced composites, which are known to be closely related with composite properties. Determination of these changes is still a tedious and challenging task because existing methods are not well developed yet. The novel method proved its ability for explaining the screw configuration effects on the attrition bore by the fibres during extrusion-compounding of plastic–matrix composites. The percentage of fibres longer than the critical length parameter was revealed to highlight the mechanical degradation of fibres during compounding. The percentage of fines exhibited the clearest correlation with the differences in fibre content of composites. Relationships found between composite tensile properties and fibre characterization parameters revealed the ability of the novel method for explaining the effects of composition and processing on composite properties.

1. Introduction

In the last decades, extensive research has been done for the use of cellulose fibres as reinforcement in polymer–matrix composites due to the advantages of these fibres, such as, low density and cost, renewability, wide availability, little damage to processing equipment, high specific strength properties, lower energy demands for processing and CO_2 neutrality.

Despite their advantages, the inclusion of cellulose fibres in a polymeric matrix brings about several problems that result in the deterioration of composite properties, and then restrict their use for end-field applications. Among these problems, the poor polymer–fibre compatibility as well as the thermal degradation and fibre damage that take place during the melt processing operations used for obtaining these composites and their end-products can be considered as the main ones.

It has been pointed out that mechanical and physical properties of fibre-reinforced composites are substantially dependent on factors determined by processing conditions, such as length distribution, final aspect ratio, dispersion, orientation, and thermal deterioration of fibres [1–3]. Thus, a main criterion for the selection of processing conditions is a compromise between the degree of fibre dispersion and attrition achieved.

Considerable research efforts have addressed the improvement of composite performance and properties through the use of compatibilizing agents for improving the hydrophilic cellulose fibres dispersion into the hydrophobic polymers used as matrices, as well as for enhancing the interfacial polymer-fibre adhesion. However, in spite of its relevant influence on composite properties, relatively less attention has been paid to the study of the cellulose fibres damage that takes place during the melt compounding and transformation processes of cellulose-reinforced composites, where the flow of suspensions of fibres into a polymer melt takes place [3]. In these processes, the cellulose fibres may suffer morphological changes due to the friction with the screws of the extruders and injection moulding machines, as well as with the tooling used and the fibres themselves [4]. The use of mild conditions (e.g., low screw rotation speeds or less kneading screw elements during compounding) may lead to a poor dispersion of the reinforcement. Conversely, aggressive compounding conditions may improve fibre dispersion as well as interfacial adhesion between fibres and polymeric matrices used, but at the expense of a greater degradation of cellulose fibres [5–13].

Several authors have studied natural fibre damage during the processing of composites. Previous work has been mainly devoted

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to polypropylene–matrix composites reinforced with different kinds of sisal and flax fibres, PLA-matrix composites reinforced with Miscanthus, bamboo or sulphite wood pulp fibres, or polycaprolactone–matrix composites reinforced with hemp fibres. Fibre geometrical parameters determination has been carried out by analysing the images captured with a digital camera or different transmission light optical microscopes and scanners [2,3,14–21]. Many of the methods used for determining the geometrical parameters of natural fibres are manual to a considerable extent, thus requiring the manual marking (e.g., with a light pen connected to a computer) of both ends of each one of the fibres to obtain the fibre length, as well as the marking of several points along the length of the fibre to estimate the mean fibre diameter. They may also require focusing properly the few fibres present at each image plane to get a sharp image suitable for the measurements. Therefore, they are time-consuming, economically not viable for routine quality controls and user-dependent, thus, there are frequent discrepancies between the geometrical parameter values obtained by different users. On the other hand, despite the much higher presence of entangled fibres than individual ones in the samples to be analyzed, measurements are limited to fibres that can be delimitated individually, because, when fibres are entangled, it becomes very difficult to distinguish which part belongs to each fibre. An in-depth comparison between the "by hand" method and an "automated method" may be found in the literature [22].

To our knowledge, despite the several works done, the determination of cellulose fibre morphological changes during the melt processing of composites as well as its relationship with processing parameters (such as, screw configuration and rotation speed, processing temperature, fibre loading or composite composition) and composite properties is still a tedious and challenging task. This is, in part, due to the inherent difficulties in determining the geometrical parameters of cellulose fibres after compounding as a consequence of their natural origin, which gives rise to a very wide range of lengths and diameters. On the other hand, progresses in developing reliable and automated methods for determining cellulose fibres geometry changes upon melt processing are still needed. This would enable for a proper design of the formulations and the melt compounding and processing technologies used for producing cellulose-reinforced composites, as well.

Therefore, this work is devoted to developing a novel automated method, which eliminates or substantially reduces the aforementioned problems of the manual methods and enables the determination of the geometrical parameters of cellulose fibres in polymer–matrix composites. In addition to the use of fluorescence optical microscopy, the novelty of this method mainly stems in the possibility of measuring the entangled fibres, the absence of restrictions regarding the measurement of long fibres and the use of a different mathematical algorithm based in the calculation of the perimeter and area of the fibres, which avoids some problems (such as the overestimation of the length and aspect ratio of fibres incurred when using an elliptic model [22]). It also enables the obtaining of accurate results in a relatively short time frame (15 min for a sample with 300 fibres compared to 10–12 h when the manual method is used). Since extrusion has been reported as the process step causing the most fibre breakage in the production of biocomposites [18], the reliability of the novel method has been assessed against its suitability for determining the changes occurring in the cellulose fibre geometrical parameters along the screw of a continuous extrusion-compounding process scaled down from an industrial one. The influence of three different screw configurations, along with the cellulose content on the fibre characterization parameters of cellulose fibres was analyzed. Finally, tensile properties of composites were analyzed along with fibre characterization parameters for trying to enlighten their relationships.

2. Materials and methods

2.1. Materials and reagents

The bleached eucalyptus Kraft pulp was supplied by ENCE-Navia, a kraft pulp mill located in Navia, Asturias (Spain). It was made from *Eucalyptus Globulus* wood by kraft pulping and subsequent bleaching using chlorine dioxide (elemental chlorine free bleaching). Its chemical composition (by weight) was: cellulose 91.88%; hemicelluloses 7.71%; lignin 0.16%; extractives 0.07%; ash 0.18%. The geometrical parameters of bleached *Eucalyptus* cellulose fibres used were: average length: 767 μm (71.2), average fibre aspect ratio: 48.9 (10.3), percentage of fines: 4.6, polydispersity index of length distribution: 1.009. Values in parentheses indicate standard deviations. Fibre length and diameter distributions of the fibres used as raw material are presented in Figs. 1 and 2.

The polypropylene used as matrix consisted of a Moplen HP 648U homopolymer (BASELL Polyolefins, Barcelona, Spain) with a density of 900 kg/m^3 (ISO 1183) and a melt flow index of 75 g/10 min, measured at 230 °C and 2.16 kg, according to ISO 1133 standard.

2.2. Composite preparation

Prior to compounding, bleached cellulose fibres were dried at 105 °C in an air-circulating oven for 3 h down to a moisture content lower than 1 wt%. Composites with 30, 40 and 50 wt% of

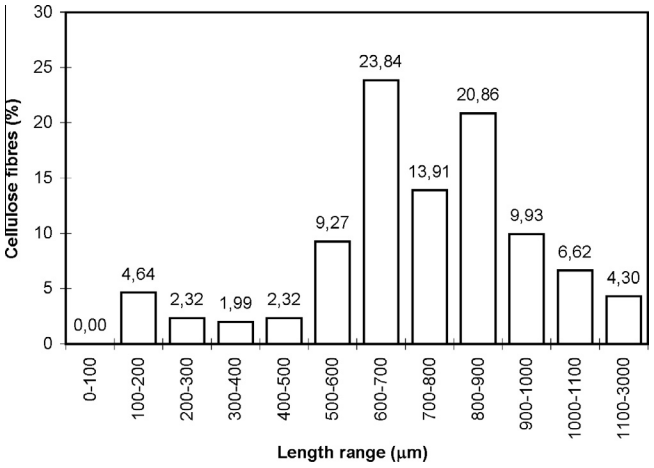

Fig. 1. Fibre length distribution of the bleached *Eucalyptus* cellulose fibres used.

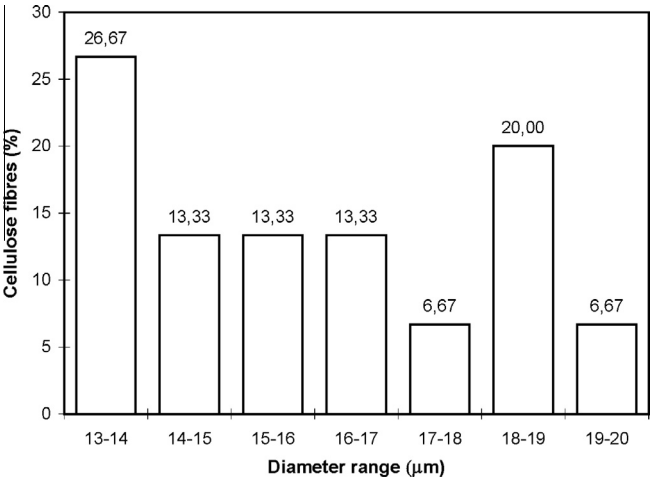

Fig. 2. Fibre diameter distribution of the bleached *Eucalyptus* cellulose fibres used.