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Evolution of lignocellulosic fibre lengths along the screw profile during twin screw compounding with polycaprolactone

Françoise Berzin [a,b,*], Bruno Vergnes [c], Johnny Beaugrand [a,b]

[a] Université de Reims Champagne-Ardenne, UMR614 Fractionnement des AgroRessources et Environnement, F-51100 Reims, France
[b] INRA, UMR614 Fractionnement des AgroRessources et Environnement, F-51686 Reims, France
[c] MINES ParisTech, Centre de Mise en Forme des Matériaux (CEMEF), CS 10207, F-06904 Sophia Antipolis, France

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ABSTRACT

Composites made of polycaprolactone reinforced by 20% hemp fibres were prepared by melt blending in a twin screw extruder (TSE). The influence of the extrusion parameters (feed rate and screw speed) on the fibre length evolution along the screw profile was investigated. The fibre length rapidly decreased after the introduction of the fibres and during the flow through the kneading blocks. Fibre fragmentation was increased at high screw speeds and low feed rates. The flow conditions along the TSE were calculated using Ludovic© software, focusing on the specific mechanical energy (SME) provided to the fibres. The fibre length evolution can be correctly estimated for various flow conditions using an exponential function of the SME.

1. Introduction

The use of lignocellulosic fibres to reinforce polymers and to design composite materials is growing due to bioeconomy requirements, the sustainability of this natural resource, its relatively low cost and its weight advantage when compared to glass or carbon fibres [1–3]. Natural fibres composites are usually produced by compounding fibres with other materials in twin screw extruders. As for synthetic fibres, the main problem is the change in fibre length (L) and aspect ratio (L/D, length/diameter), due to the high stresses encountered during the process. Individual fibres are generally cemented together into bundles, and then into a group of bundles, by a matrix that consists of several amorphous polymers [4,5]. The defibrization process concomitantly results in decohesion (when a fracture occurs in the interfibre cement) and in fragmentation of the fibre elements. Fragmentation occurs when the intrafibre cell wall is damaged, which is comparable to the breakage that occurs in synthetic fibres (i.e., glass fibre). During the extrusion process, both decohesion and fragmentation occur in the composite material, leading to fibre individualization and length reduction [6–8]. Contrary to rigid glass fibres, whose rupture mechanisms are well known and modelled [9–12], lignocellulosic fibres are generally flexible and can entangle to form large aggregates. Novel insights about flax fibre ruptures were recently provided through direct observations using rheo-optics [13]. Rupture by fatigue was observed, and the authors confirmed that natural defects (“kink bands”) initially present on flax fibres strongly affect the rupture location. The influence of thermomechanical conditions on the fibre cell wall properties [14] and on the fibre length and aspect ratio have been studied with internal mixers [15] or TSEs [16–20]. In a previous study [20], we demonstrated that differences in defibrization affect the fibre L/D ratio, which in turn influences the mechanical properties of the final composite. This phenomenon was also observed by Jiang et al. [21]. Regarding the process parameters, the final fibre length was reduced with decreasing feed rates and increasing screw speeds. In contrast, the L/D ratio was less affected by changes in the above-mentioned processing conditions, indicating a simultaneous reduction in fibre length and diameter. The fibre length and aspect ratio could be roughly evaluated for different processing conditions using an exponential function of the specific mechanical energy, as suggested for glass fibres by Inceoglu et al. [10].

In this paper, we describe the development of a predictive lignocellulosic defibrization model by following the length evolution along the screws during TSE compounding. This work involved the same polycaprolactone matrix, lignocellulosic fibre (hemp) and TSE materials as described in a previous study [20]. We characterized the evolution of the fibre length distribution along the screw profile for different processing conditions and established a theoretical model describing these results.

* Corresponding author at: Université de Reims Champagne-Ardenne, UMR614 Fractionnement des AgroRessources et Environnement, F-51100 Reims, France.
E-mail address: francoise.berzin@univ-reims.fr (F. Berzin).

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2. Experimental

2.1. Materials

The matrix consists of polycaprolactone (PCL), selected for its low melting temperature (60 °C), which is of interest when compounding lignocellulosic fibres. PCL (Capa© 6800), provided by Perstorp (United Kingdom), has a molecular weight of 80,000 g/mol and a melt flow index (MFI) of 3 g/10 min (160 °C, 2.16 kg). At 160 °C, PCL is quasi-Newtonian with a viscosity of 230 Pa s, as shown in Fig. 1.

The fibres used in this study originate from hemp stems (*Cannabis sativa L*, variety: Fedora 17) grown in Aube (France) in 2009 and were supplied by Fibres Recherche Développement® (Troyes, France). To minimize the heterogeneity and the length dispersion, kilograms of long scutched bast fibres were manually hacked and chopped into a homogeneous masterbatch approximately 1.4 ± 0.34 cm in length. Initial average diameter of the hemp fibres was around 300 μm, leading to a L/D ratio of 47. The fibre samples were preconditioned at 50% relative humidity in a climatic chamber before testing.

2.2. Extrusion conditions

The composites were prepared using a laboratory-scale TSE Clextral BC 21 (Firminy, France). The extruder has a diameter (D) of 25 mm and a length (L) of 900 mm (L/D ratio: 36). The screw profile used is shown in Fig. 2. The TSE is composed of a block of kneading discs with staggering angles of −45° for melting, followed by a second one with angles of 90° and then by a third with angles of −45° (Table 1). PCL and hemp fibres can be introduced either simultaneously in the hopper (barrel 1, before the melting zone) or in two steps (PCL in barrel 1 and all the fibres in barrel 4, after PCL melting). In all cases, a venting zone for eventual water steam evacuation is located in barrel 7.

The barrel temperature was set at 100 °C, and the experiments consisted of varying the flow rate (1, 2 and 3 kg/h) at a constant screw speed (200 rpm) and varying the screw speed (100, 150, 200 and 300 rpm) at a constant flow rate (2 kg/h), as described in Table 2. The fibres were mixed with PCL at a concentration of 20 wt.%. The fibres were fed manually to prevent possible irregular feeding, which usually occurs with long fibres when using either volumetric or gravimetric feeders. Before the experiment, a lot of beakers were prepared, with 2 g of fibres in each of them. During the extrusion, they were poured each 15 seconds into the feeder or the barrel opening. It was checked that this methodology ensured a very stable and constant feeding. After reaching steady-state conditions, the feeding and screw rotation were suddenly stopped, the barrel was cooled by water circulation and the barrel was extracted. The samples were then obtained from the screws at different locations, as indicated in Fig. 2 (positions 1–6: fibres are introduced before the melting zone; positions 4–6: fibres are introduced after the melting zone).

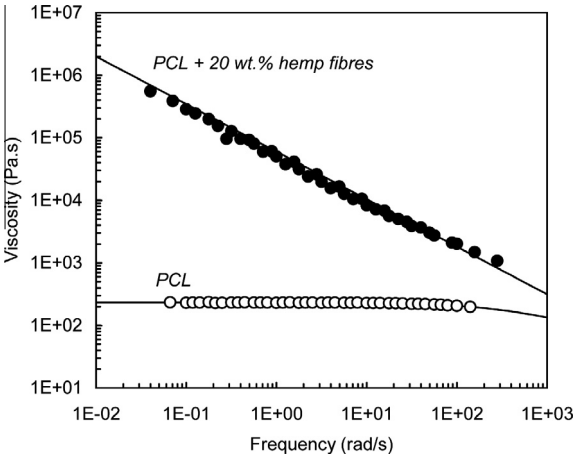

Fig. 1. Viscosity curves of PCL and composite with 20 wt.% fibres at 160 °C. Symbols are experimental points, full lines are fits by a Carreau–Yasuda law (PCL) or a power law (PCL + 20 wt.% hemp fibres).

2.3. Flow modelling

To estimate the parameter values that cannot be measured experimentally during the compounding process, we used Ludovic© flow simulation software dedicated to TSE [22]. Specifically, Ludovic© was used to calculate the specific mechanical energy (SME) transmitted to the composite all along the extruder and to predict the average length evolution during the compounding process.

2.4. Composite characterization

After extrusion, the samples were analyzed to quantify the fibre length distribution. First, the PCL matrix was extracted using the Soxhlet methodology [20] to determine the fibre content. This technique was employed to evaluate the possible fibre content variation during compounding and to verify that the average value satisfied the 20% target.

The fibre dimensions and morphologies were further obtained via image analyses using optical microscopy and MorFi software (Techpap, Grenoble, France). A mass of approximately 0.6 g was diluted in water extemporarily, and the fibres were tested in duplicate or triplicate samples to determine their mean values. Over 100,000 elements were counted for each sample. By convention, elements having a length of >200 μm were considered fibres, while shorter elements were considered fines. This limit is imposed by the MorFi system. It corresponds roughly to the length under which the mechanical reinforcement is less interesting. The fibre elements were further characterized by their aspect ratio L/D. However, elements longer than approximately 10 mm cannot be analyzed with this system due to technical restrictions. Fortunately, these long fibres remained seldom in the extruded samples. Moreover, although optimized dilution procedures were used, when agglomerates of entangled long fibres were present, they could not be taken into account via the employed method. In this case, the calculated distribution was underestimated, and the values were not used in the discussion.

3. Results and discussion

3.1. Extrusion conditions

First, focusing on the extrusion conditions that define the thermomechanical treatment received by the hemp fibres during the compounding process is necessary. Indeed, a temperature of >160 °C has been reported to affect fibre properties [20], and the conditions (e.g., inert atmosphere, presence of oxygen, temperature ramp and time of exposition) can modify the thermal degradation temperature [23]. Moreover, we previously demonstrated the relationships between the SME and fibre dimensions [20]. Fig. 3 shows the final product temperature variations, measured at the die exit, and the global specific energy, deduced from the extruder torque, as functions of the screw speed. With these results, we observed a linear increase in both parameters with increasing screw speed. The maximum temperature is 158 °C, which should prevent