Effect of the Preparation of Cellulose Pellets on the Dispersion of Cellulose Fibers Into Polypropylene Matrix During Extrusion

Marie Le Baillif, Andreas Echtermeyer

Department of Engineering Design and Material, Norwegian University of Science and Technology, Richard Birkelandsvei 2b, Trondheim NO-7491, Norway

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ABSTRACT: Cellulose fibers are rarely used for the extrusion of composites because of the problems in feeding them into the extruder and in dispersing them properly. Pelletization made it possible to feed cellulose fibers into extruder, but it reduced dramatically the fiber length. The goal of this study was to optimize the pelletization process for extrusion applications. Bleached sulfite cellulose fibers were pelletized at different moisture contents and with the optional addition of carboxymethyl cellulose (CMC). The pellets were subsequently extruded with polypropylene matrix without compatibilizer. Fiber dispersion and fiber breakage during extrusion were investigated. Pre-blending of polymer and fiber pellets and introduction of the fibers through a side extruder were compared. CMC acted as a processing aid during pelletization, resulting in lower fiber breakage but in compact and stiff pellets. Lower moisture content also increased the compactness of the pellets. The dispersability of the fibers during extrusion decreased with increased pellets' compactness. CMC created inter-fiber bonds, decreasing further the fibers' dispersability. The fiber length in the composites was the same regardless of the pelletization parameters. Early introduction of the fibers improved fiber dispersion. Feeding through side extruder was more stable and more reliable than pre-blending. © 2009 Wiley Periodicals, Inc. J Appl Polym Sci 115: 2794–2805, 2010

Key words: cellulose fibers; pelletization; polypropylene; extrusion; composites

INTRODUCTION

Cellulose fibers polyolefin composites were extensively studied during the last years.[1–14] Those studies were mainly presenting the mechanical properties of composites which were often prepared using batch processes like thermo-kinetic mixers[1,10,13–15] or mixing chambers.[2,6,7] Only few studies reported the use of an extruder for the compounding of cellulose fibers polyolefin composites,[9,11] mainly because of problems of handling the cellulose fibers.[6,9] Cellulose fibers, but also natural fibers in general, are known to have an aspect ratio (length/diameter) over 50, to be fluffy, to have low bulk density and therefore to show no free flowing characteristics. These properties of cellulose fibers resulted in difficulties in doing, controlling their transportation and also their feeding into extruders.[16] By comparison, wood flour has lower aspect ratios (typically up to 10 only), higher bulk density and can be handled as a powder.[16]

In recent studies, the handling of cellulose and natural fibers was improved by pelletizing or granulating the fibers.[17,18] Both names "pelletization" and "granulation" were used for similar processes depending on the authors or to differentiate two processes. Warnes et al.[16] have patented a method to prepare "granules" of wood fibers by compression molding of impregnated wood fiber before granulation. Sears et al.[17–19] made "pellets" of never dried cellulose fibers before extrusion with polyamide. The never dried pulp, mixed with some chemicals, was pushed through a die plate by rotating roll-mills. They compared the "pelletization" process with the "granulation" which, there, consisted in cutting paper sheets using a rotary knife cutter.[18] Pelletization and mainly the granulation method were found to reduce the average fiber length. Based on the same pelletization method, we have shown in previous work that "pelletization" allows consistent feeding of cellulose fibers into extruder. However, it was also found that the average fiber length was drastically reduced by 50%, limiting the reinforcement potential of the fibers.[20,21]

Nowadays, the pelletization or granulation method is used for many applications such as the production of wood pellets for fuel, animal bedding and animal food, but also for the preparation of tablets. Granulation is defined to be "the process of collecting particles together by creating bonds between them by compression or using a binding agent."[22]

Correspondence to: M. L. Baillif (marie.lebaillif@ntnu.no).

Four mechanisms were reported to bind particles within pellets: (a) solid-solid interactions, (b) capillary forces in a liquid/air/solid system, (c) adhesive and cohesive forces between particles and binders and (d) mechanical entanglements of the particles.[23] In the case of wood pellets, the lignin is acting as binding agent. During processing, the temperature can increase up to 130°C making the lignin in the wood more plastic.[24] In the case of cellulose fibers pellets, most probably mechanisms to built networks might be hydrogen bonding between fibers and mechanical entanglements. Hydrogen bonds are known to be very strong between cellulose fibers. Moreover, it has been shown that hornification (which describe the irreversible structural changes in pulp fibers during drying[25]), caused by the irreversible coalescence of cellulose microfibrils, is occurring during drying and rewetting cycles, leading to irreversible bonding between cellulose fibrils and to the stiffening of the fibers.[26,27] The hornification of pulps has been shown to be mainly dependant of the number of drying-remoistening cycles and to especially occur during the first cycles.[25] Hornification could be of importance for the determination of the properties of cellulose fibers pellets.

In the preparation of cellulose fibers pellets, Sear et al.[17–19,28] used carboxymethyl cellulose as a processing aid. Carboxymethyl cellulose (CMC) is a cellulose ether widely used nowadays under trade names like Aqualon®, Blanose® or Bondwell® among others. Its applications are very wide from the food industry to the paint, paper, oil industry but also in the pharmaceutical industry as thickener, rheological modifier, gelling agent, foam stabilizer, lubricant, etc. CMC is reported to be used in the paper industry as a wet-end additive for surface sizing and as a component for coating.[29] More recently, CMC has been found to be absorbed on the surface of cellulose kraft fibers and that this adsorption would be partially irreversible.[30,31] It was proposed that this irreversible adsorption could be due to co-crystallization on the nonsubstituted part of the CMC structure with the cellulose fibrils on the fiber surface which have the same structure. The investigation of the cellulose pellets structure should give better understanding of the behavior of the pellets during the process for compounding composites.

Earlier studies have shown that cellulose fibers do not properly disperse within polyolefin matrices, because of incompatibility of hydrophilicity.[4,5] In a recent work, the dispersability of cellulose fibers into polypropylene matrices was found to be improved by increasing screw speed during extrusion or by extruding twice the composite materials.[21] It was also found that improvements of dispersion were accompanied by large fiber breakage and partial thermal degradation of the materials during extrusion. It is well known that good fiber dispersion and long fibers are more suited for obtaining composites with high mechanical properties.[4,9,10,32,33] It is expected that more knowledge about the preparation of the cellulose pellets could allow the production of pellets of cellulose fibers that would be more suited fore extrusion. Such pellets should contain fibers as long as possible r to keep their potential of reinforcement, meaning that fiber breakage during pelletization should be the lowest as possible. Moreover, such pellets should be easily dispersed into polyolefin matrices during extrusion, and they should be easily handled, transported and stored.

The goal of this work has been to optimize the pelletization of bleached sulfite cellulose fibers from softwood in view of the extrusion of composite materials. Therefore, fiber breakage during pelletization was reduced by optimizing moisture and CMC contents during processing. Moreover, the ability of the fibers to be dispersed during compounding was analyzed depending on (a) the formulation of the pulp during pelletization, (b) the nature of drying of the cellulose pellets after pelletization, and (c) the way of introducing the fibers pellets into the extruder. Finally, the need of pelletizing cellulose fibers prior extrusion was investigated.

EXPERIMENTAL

Materials

Cellulose fibers: Paper sheets of 70 × 80 cm^2 of bleached sulfite cellulose fibers from softwood (grade derivate MCC) were supplied by Borregaard Chemcell AS (Sarpsborg, Norway). Those fibers contained 95% cellulose, 3.5% hemicelluloses and some extractives.

Additive for pelletization: Carboxymethyl cellulose (CMC) from Noviant Oy (Äänekoski, Finland) was used as processing aids. CMC was supplied as powder.

Polymer matrix: The polymer matrix was a polypropylene (PP) block copolymer BE170MO supplied by Borealis AS (Stathelle, Norway). The melt flow index (MFI) of the PP was 13 g/10 min and the density was 0.902 kg/m^3.

Processing aid for extrusion: A lubricant Struktol TPW 709 purchased from Struktol Compagny of America (Stow, Ohio, US) was added during the extrusion of the composite materials.

Processing

Pelletization

For each batch, the sheets of paper were cut by hand into small pieces before storing for 2 days in a plastic bag. Water was added inside the bags to achieve

TABLE I
Nomenclature of the Materials: Pellets and Composites

	Pelletization			Extrusion
Material	Dry content [wt %]	CMC content[a] [wt %]	Drying	Feeding method
25-0-PB	25	0	Air	PB
25-3-PB	25	3	Air	PB
40-0-SE	40	0	Air	SE
40-0-PB	40	0	Air	PB
40-3-PB	40	3	Air	PB
40-0-OV-PB	40	0	Oven	PB
40-3-OV-PB	40	3	Oven	PB
60-0-PB	60	0	Air	PB
60-3-PB	60	3	Air	PB
F-0-PB	–	0	–	PB

[a] CMC content defined by comparison with the weight of dry pulp. PB, pre-blending; SE, side extruder; OV, oven dried.

either a moisture content of 50 wt % or the desired moisture content if it was over 50 wt %. The wet pieces of paper were then fluffed three times to obtained pulp suitable for pelletization using an in-house built fluffing equipment consisting of a fast rotary unit. The wet pulp was mixed with water to adjust the moisture content of the pulp and with CMC into a Hobart Mixer for 4 min. The composition of the wet pulp before pelletization is presented in Table I. Cellulose fibers pellets were produced using a Kahl pellet press (Kahl GmbH & Co. 14–175, Reinbek, Germany). During the pelletization, the pulp was pressed through a die plate (holes of 4 mm diameter) by two rotating roll mills at a speed of 16.5 rpm. A rotary knife was cutting the threads of pulp below the plate, see Figure 1. The wet pellets were finally dried either by air or in an oven. Oven dried specimens have the code "OV" in this paper. The materials are referred in the study by their dry content during pelletization.

Flocks preparation

As an alternative to pelletization, paper flocks were prepared to introduce longer fibers into the extruder. Sheets of paper were cut in strands of about 1 cm wide using a knife. The strands were then cut in flocks of about 3 mm long using a roll mill knife before storage.

Compounding

The cellulose materials were dried overnight in an oven at 80°C to obtain a moisture content below 1 wt % during the compounding of the composite materials. Composite materials containing 20 wt % fibers were manufactured using a twin screw co-rotating extruder ZSK 25 WLE (Coperion Werner and Pfleiderer, Stuttgart, Germany). The processing temperature ranged from 175 to 200°C, see Figure 2. The out put was fixed at 7.5 kg/h and the screw speed at 300 rpm. The polymer matrix was obtained by pre-blending the PP and the lubricant with a weight ratio of 20 : 1. Ventilations were used in zones 6 and 8. Vacuum was used in zone 10 to remove volatile compounds. The polymer matrix was introduced in zone 1 using a K-tron gravimetric feeder (Niederlenz, Switzerland). The cellulose pellets were either pre-blended (coded PB in this paper) with the polymer matrix and therefore introduced in zone 1, or introduced separately using a second gravimetric feeder in the zone 4 of the extruder through a side extruder (coded SE). The different formulations of the composites materials are shown in Table I. The melted composites were recovered at the exit of the extruder and directly compression molded to make plates of $10 \times 6.8 \times 0.1$ cm^3. The compression molding cycle was: first 1 min at 180°C without pressure, then 4 min at 180°C with a pressure of 6 MPa, and finally cooling at 50°C for 2 min under a pressure of 12 MPa.

Characterization

Apparent density

Paper sheets: The apparent density was measured according to standard ISO 534 on sheets of area $A = 20 \times 20$ cm^2. The thickness of the paper sheets was measured using a L&W Micrometer 51 (Lorentzen & Wettre, Stockholm, Sweden). The specimens were stored for 2 days at normal condition of humidity and temperature (23°C and 50% humidity) before measurement. Five measurements of the thickness were done for each sheet of paper to reduce inaccuracies due to the roughness of the paper. The results presented correspond to the average apparent density obtained from 20 specimens. The

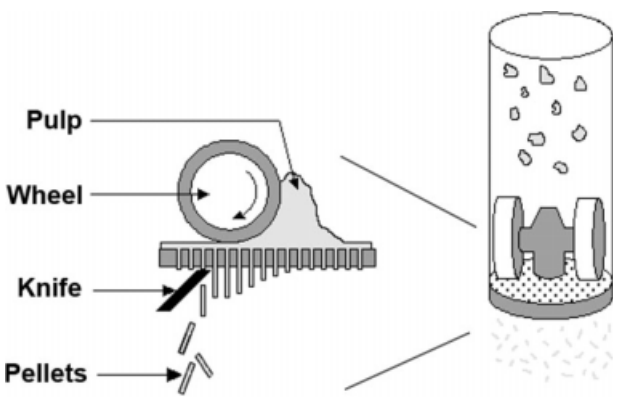

Figure 1 Pelletization process.

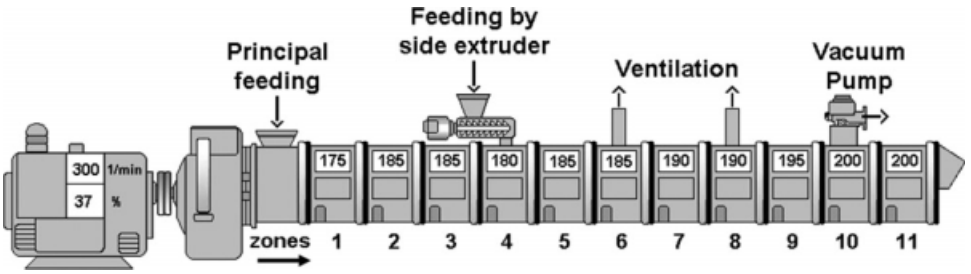

Figure 2 Extruder and compounding parameters.

apparent density is obtained by the eq. (1) (where ρ_{app} is the apparent density in g/cm^3, w the weight of the paper sheet in g and t the thickness of the sheet in cm).

$$\rho_{app} = \frac{w}{t \times A} \quad \text{(Eq. 1)}$$

Other materials: The apparent densities of polymer pellets, cellulose fiber pellets, fluffed cellulose pulp, and paper flocks were measured by adapting the standard method ASTM B212-99. After 2 days storing at normal condition of humidity and temperature (23°C and 50% humidity), the tested material was filled free falling into a cylinder of 1.051 L (about 13.5 cm diameter to limit border effects). The level was carefully flattened and the full cylinder was weighted. The apparent density was then calculated using eq. (2) (where ρ_{app} is the apparent density, w_f the weight of the full cylinder, w_e the weight of the empty cylinder and V the volume of the cylinder). The presented results were the average of 20 repeated measurements. The standard deviations were very small (about 0.004 g/cm^3) and were hardly visible on the graph.

$$\rho_{app} = \frac{(w_f - w_e)}{V} \quad \text{(Eq. 2)}$$

Pellet dimensions

Dimensions of the cellulose pellets were examined by sieving 100 g of pellets of each formulation. The sieving was performed using a shaking machine and a column of 6 sieves (5/16, 3 1/2, 5, 7, 12, and 16 mesh, with holes of 8, 5.6, 4, 2.8, 1.7, 1.18 mm, respectively) for 1 min and 30 s at a shaking speed of 300 rpm. The fines were recovered in a plastic bag. After shaking, each sieve was weighted carefully. By being sieved, the pellets will be classified according their smallest dimension. If the pellets had cylindrical shape, it would correspond to their diameter. If the length of the pellets was approaching the diameter of the pellets, the pellets were most probably having a spherical shape. The smallest dimension would then be their diameter.

Fiber dimensions

The fiber dimensions were measured using the automatic fiber quality test equipment Fiber Master (Lorentzen & Wettre, Stockholm, Sweden). For each measurement, 10,000 to 12,000 fibers were measured based on image analysis.[34] Only fibers having an aspect ratio (length/diameter) over 4 were detected. The dry fibers were dispersed in water before measurement. To analyze the fibers contained in the composite materials, the PP matrix was removed by extraction using p-xylene at 150°C for 4 days.

Fiber dispersion

The fiber dispersion in the composite materials was investigated using the compression molded plates. A frame was placed over of the plate which was then placed between a light source and a camera. For each material, several pictures of several plates were taken to have representative data. Each picture was then analyzed using the ImajeJ program.[35] The scale of the picture was first measured. Then the picture was transformed to be in gray scale and the background was subtracted before adjusting the threshold. At that stage, fiber agglomerates appeared as black dots, which were measured and counted using the shape descriptor plugging of the ImajeJ program.[36] The presented results are the average of a minimum of 10 pictures for each material. To allow comparison and because of differences in thickness between the plates, the results are given per volume unit of composite material.

Morphology

The morphology of cellulose fibers pellets was analyzed using a Hitachi S-3000N scanning electron microscope (Hitachi High Technologies America, Pleasanton, CA) with an acceleration voltage of 5 kV. Gold was sputtered on the analyzed surfaces to avoid charging. The discussion presented in this study is based on the analysis of several pellets. However, only the extreme cases are presented in the Figures.

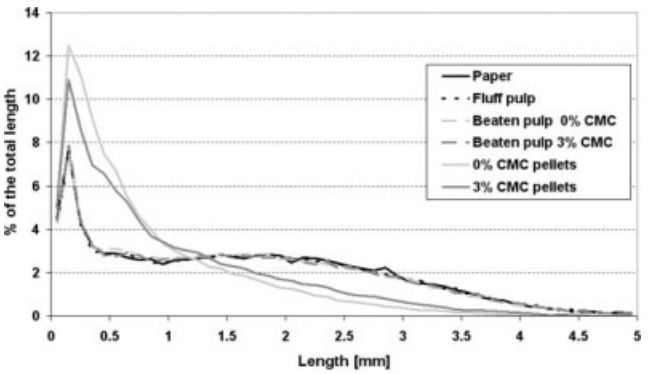

Figure 3 Fiber length distribution at all the steps of the pellets preparation.

RESULTS AND DISCUSSION

Pelletization

Process ability

Pelletization was only possible for pulp having a dry content between 25 and 60 wt %. For pulps with a dry content lower than 25 wt %, the water could not be absorbed and it dripped out. Pulps having a dry content over 60 wt % were not able to flow though the holes of the plates. When water was added to the cellulose pulp, the hydroxyl groups of cellulose formed hydrogen bonds with water rather than with hydroxyl groups of other cellulose fibers. As a result, the inter fiber interactions were lowered and the fibers gained mobility. For very dry pulp, the fiber interactions were too strong to allow sufficient mobility of the fibers to be pelletized. Heat was produced during the pelletization as an indication of the shear forces applied during processing. The heat was found to increase with the dry content of the pulp. The wetter the pulp was, the looser was the cellulose fiber 3D network, giving less friction during pelletization and subsequently producing less heat. In addition, water has a high heat capacity, requiring more heat energy to warm up a very wet pulp. The addition of CMC significantly improved the flowing of the pulp during the pelletization, decreasing also the heat produced during processing. However, this improvement was not sufficient to make possible the pelletization of pulp with a dry content over 60 wt %.

Best suited dry content of cellulose fiber pulp for performing the pelletization was around 40 wt %. An optional addition of CMC would facilitate the processing but would not be required.

Fiber breakage

Figure 3 shows the length distribution of the cellulose fibers at all the steps of the preparation of the pellets. It is clear from the Figure that the pelletization step was the only one affecting significantly the fiber length, leading to shorter fibers especially when no CMC was used. The average length of cellulose fibers was found to be reduced from about 1.5 – 1.65 mm to 0.78 – 0.94 mm during pelletization, see Figure 4. The fiber breakage was most probably due to shear applied during the pelletization. This energy was dissipated within the fiber breakage but also by the production of heat. As visible in Figure 4, the fiber length reduction was not affected by the dry content during pelletization. However, the addition of CMC lowered the fiber breakage. As mentioned above, CMC acted as a lubricant during the pelletization, reducing the frictions during processing. It is then logical that less shear energy was produced when CMC was added, limiting the fiber breakage during processing. The interactions between CMC and cellulose fibers explaining the lubricant effect of the CMC will be described in more details in the next section.

Pellets' morphology

For all the materials, the pellets had a cylindrical shape. Figures 5 and 6 show the cellulose fibers pellets of 25-0-PB and 60-3-PB, respectively (for an explanation of the pellets codes see Table I). CMC addition and higher dry content during pelletization made the pellets more compact and improved the cohesion of the pellet. The entanglements between the fibers seemed to be loosened for 25-0-PB pellet, see Figure 5(a,b), while the fibers were much tightened for the 60-3-PB pellet, see Figure 6(a,b). The fibers are clearly visible on the surface. It is noticeable that the fibers tend to be aligned in the flow direction of the pellets. However, the alignment is more visible when CMC was used and with higher dry content while pelletizing, see Figure 5(a), and 6a. The pellet 25-0-PB shows many fibers in the

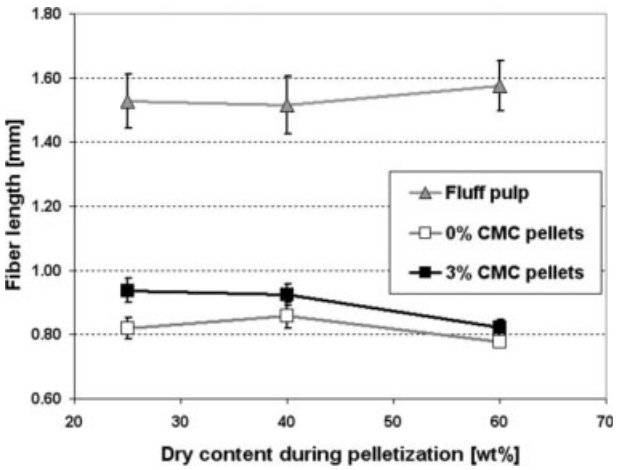

Figure 4 Average fiber length in pellets and fluff pulps.

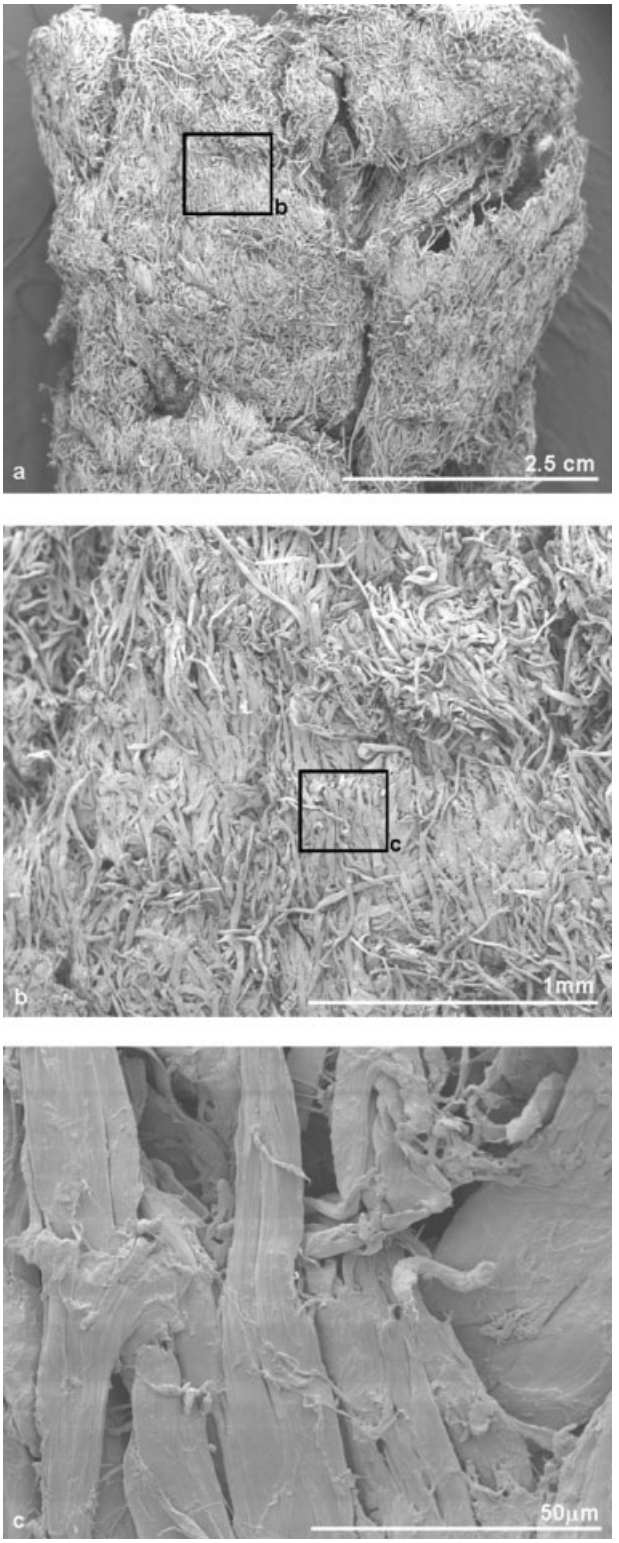

Figure 5 SEM picture of 25 – 0 – PB pellet (see Table I): magnification a: ×18, b: ×50 and c: ×1000.

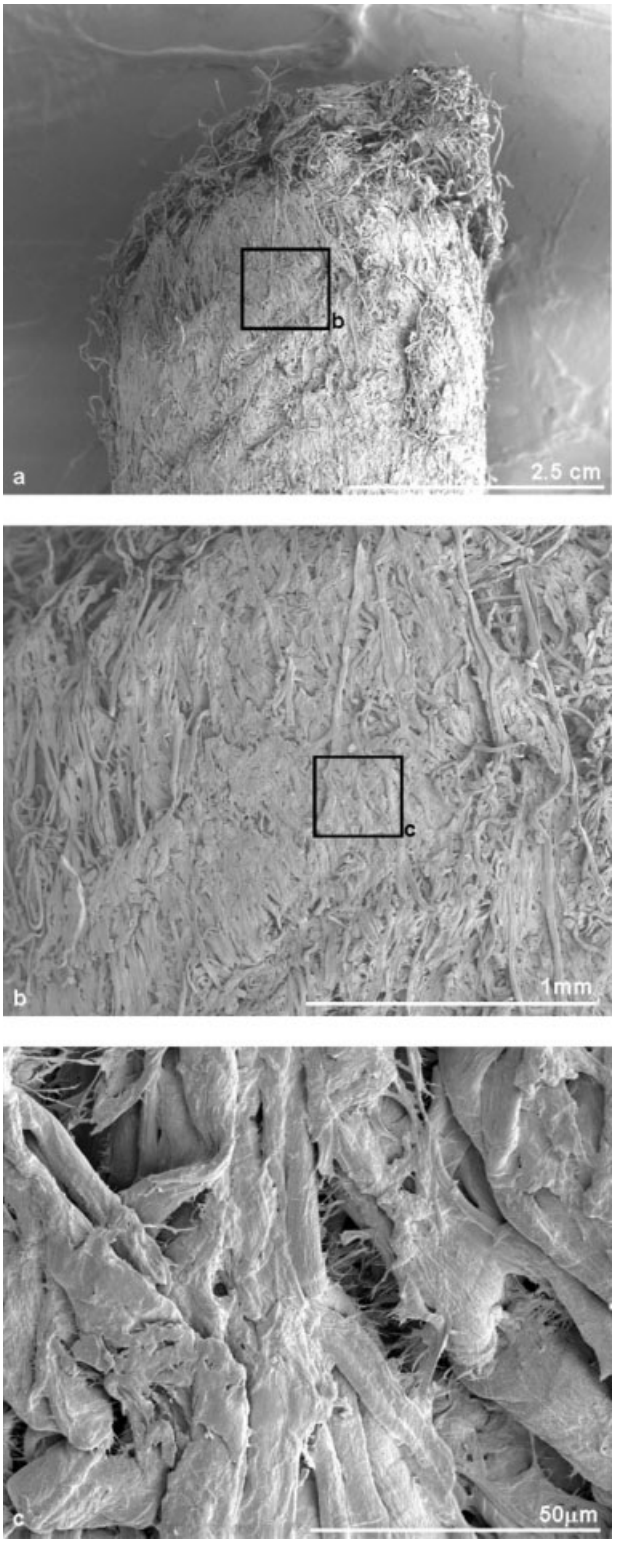

Figure 6 SEM picture of 60 – 3 – PB pellet (see Table I): magnification a: ×18, b: ×50 and c: ×1000.

transverse direction whereas the pellet 60-3-PB shows almost no fiber in the transverse direction, See Figures 5(a,b) and 6(a,b), respectively. The pellets containing CMC seem to show some adhesion between the fibers, see Figure 6(c) whereas the pellets without CMC show fibers clearly distinct from each others, as visible on Figure 5(c).

The cohesion and the compactness of the pellets seemed to be defined by the degree of fibers entanglements into the 3D network. This could be

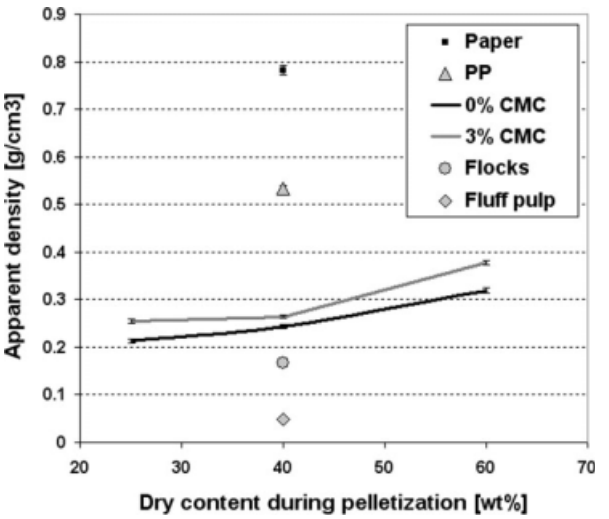

Figure 7 Apparent density of the materials.

explained by the pelletization process. The cellulose fibers are known to have strong fiber-fiber interactions through hydrogen bonding, creating a 3D network.[4,33] This network can be loosened by the presence of water which can penetrate the fibers. The hydrogen bonds are then changing from fiber-fiber to fiber-water. When CMC is added to the wet pulp, it is absorbed on the fiber surface.[30,31] The adsorption is believed to occur with the non substituted parts of the CMC molecules which are cellulose. This is a partially irreversible process because co-crystallization with the cellulose of the fibers can occur. However, CMC molecules contain anionic parts which create repulsive forces with the negatively charged cellulose fibers. Hence, the addition of CMC is breaking the cellulose fiber 3D network, allowing high mobility of the fibers and therefore allowing possible alignment of the fibers during the pelletization. It seems logical that the CMC had the most significant effect on the mobility of the fibers.

The most compacted pellets were the ones made at high dry content (60 wt %). This might be explained by the concentration of CMC at the fiber surface. For the two materials (25-3-PB and 60-3-PB) the dry fiber-CMC ratio was the same, but the amount of water was different. It is believed that the CMC was diluted by the water, since CMC is known to be water soluble, and therefore the concentration of CMC at the fiber surface was lower for 25-3-PB than for 60-3-PB. As a result, the adsorption was less important, leading to lower repulsion forces between fibers having absorbed CMC. During the drying, the water was removed and co-crystallization between cellulose fibrils but also between CMC and cellulose fibrils on the fiber surface occurred. Because the CMC was added on the wet pulp, the CMC chain might be able to co-crystallize with different fibrils and eventually with different fibers. This could explain the visible adhesion on the high magnification pictures for the CMC added materials [Fig. 6(c)].

Apparent density and pellets dimensions

Figure 7 shows the different values of the apparent density measurements. Cellulose pellets had an apparent density between 0.21 g/cm^3 for 25-0-PB pellets and 0.38 g/cm^3 for 60-3-PB pellets. Sears et al. measured an apparent density of cellulose pellets 0.6 g/cm^3.[19] By analyzing the same pellets as Sears et al., the apparent density was measured to be 0.22 g/cm^3 (standard deviation of 0.004 g/cm^3), which is similar to the other cellulose pellets analyzed in this study. However, the large difference between the measurements presented by Sears et al. and those presented in this work, shows clearly that this method can only be used for comparison between the materials. The materials ordered from the higher to the lower apparent density were cellulose paper > PP pellets > cellulose pellets containing CMC > cellulose pellets without CMC > paper flocks > fluff pulp. The high apparent density of 0.78 g/cm3 obtained for the paper sheets was expected since the paper sheets could be considered as a bulk material. The apparent density of the pellets was increasing with the dry content of the pulp during pelletization. The increase of apparent density with higher dry content and addition of CMC is in complete agreement with the observation made using SEM and the increase of the compactness of the pellets. The drying had no effect on the apparent density of the pellets.

The sieving of the cellulose fiber pellets showed that the major fraction of the pellets had their smallest dimension between 4 and 5.6 mm, see Table II, which corresponds to the diameter of the holes of the plate in the pellet press. This would mean that most of the pellets had a cylindrical shape. The pellets whose diameters were smaller than 4 mm may in fact correspond to fragments of pellets. Pellets with a diameter larger than 5.6 mm or 8 mm may be agglomerates of pellets. It is noticeable that only pellets pelletized at 25 wt % dry content showed pellets bigger than 8 mm. Moreover, the proportion of

TABLE II
Pellet Diameter Obtained by Sieving – Weight Percentage

	Hole sizes [mm]						
Material	8	5.6	4	2.8	1.7	1.18	0
25-0-PB	1	2	52	25	14	4	2
40-0-PB	0	0	60	22	12	3	3
60-0-PB	0	0	72	17	8	2	2
25-3-PB	1	3	83	8	4	1	0
40-3-PB	0	2	87	8	3	1	0
60-3-PB	0	0	93	5	1	0	0

pellets with a diameter between 5.6 mm and 8 mm was increasing with increased moisture content during pelletization (so lower dry content) and also with the addition of CMC. When the pellets were still wet, they could re-agglomerate before the drying. The wetter the pellets were the heavier they were, and therefore, the easier it was to agglomerate with other pellets because of gravity. By adding CMC, the water was more like gel, which could stick the pellets together. Then during the drying, inter fiber bonds were formed, leading to bigger pellets if the contact surface between the pellets was sufficient (large surface contact leading to more numerous interactions between the pellets). The size distribution of pellets prepared without CMC was quite wide, whereas the distribution of the pellets prepared with CMC was much narrower on the 4 mm pellets. When the pulp was pelletized, the threads below the plates were supposed to be cut by the knife. However, if the fiber network was loosen enough (as by the presence of water) and the pulp thread not compact enough, the gravity was sufficient to make the thread or pieces of it falling before the knife to cut them, resulting in smaller pellets. Therefore, the obtained results are in agreement with the compactness of the pellets observed above. The drying method had no effect of the pellets size.

Extrusion

Processability

It was found impossible to use the side extruder for the introduction of the flocks and the pellets which contained CMC. The screw of the gravimetric feeder could not transport the pellets. The pellets and the flocks were too large to fit in the free space allowed by the screw in the barrel of the gravimetric feeder, and they were too stiff to be broken or to have their shape changed by the screw in the barrel. It resulted in the stop of the feeder. This problem could probably be solved by changing the screw of the feeder. There was, however, no problem for introducing pellets which did not contain CMC using the gravimetric feeder and the side extruder. This shows that the addition of CMC during pelletization made the cellulose pellets stiffer. The higher hardness of the pellets containing CMC was most probably due to the high compactness and the numerous inter fiber bonds as discussed above.

There was no apparent difference in the materials due to the motor torque or the melt pressure at the die. During a period of 30 min to 1 h, the fiber content within the materials was found to be around 15 wt % for all the materials expect 40-0-SE and F-0-PB. After this delay, the composites which were prepared using pre-blending showed an increase of the fiber content. For 40-0-SE, the fiber content was found to be accurate and stable at 20 wt % during the whole processing time, while the fiber content for the flocks was found to be unstable. This might be due to segregation in size and weight in the feeder. When granules having different sizes and density are mixed together, the granules having the smallest size and the higher density go in the bottom of the feeder.[33] The obtained unstable fiber loadings could therefore have been expected since the cellulose pellets were found to have a lower apparent density and a larger size than the PP pellets as explained above. The segregation problem was avoided by the control and the regulation of the feeding rate obtained when a side extruder was used. This shows that feeding cellulose pellets through a side extruder should be preferred to pre-blending to assure consistent and regular fiber loading levels.

A slight yellowing of the composite material was noticed when CMC was present in the cellulose pellets. The yellowing might be due to the CMC which might be less thermally stable than the cellulose.

Dispersability of the pellets

Figure 11 shows plates of composites made with pellets prepared at different CMC and dry content. Dark spots were visible in all the materials corresponding to fiber agglomerates, meaning that the cellulose fibers were poorly dispersed into the PP matrix, as expected. The number and the size of the agglomerates increased with increasing dry content during pelletization, see Figures 8 and 9. The

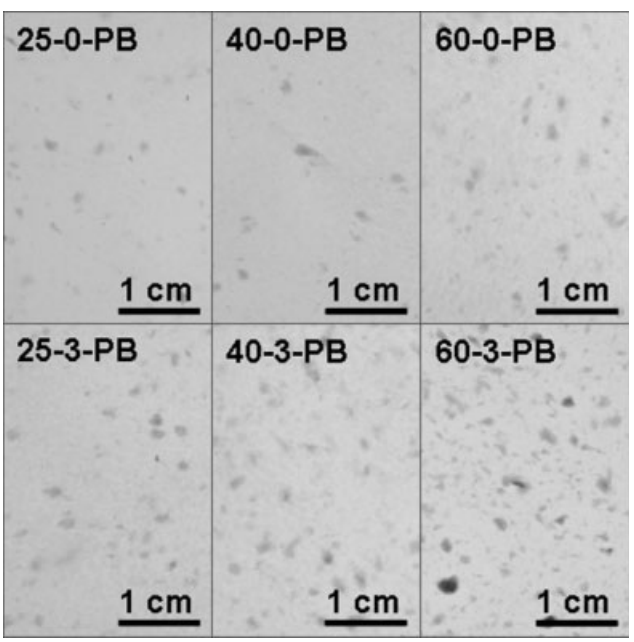

Figure 8 Agglomerates in composite materials. Effects of CMC and dry content during pelletization.

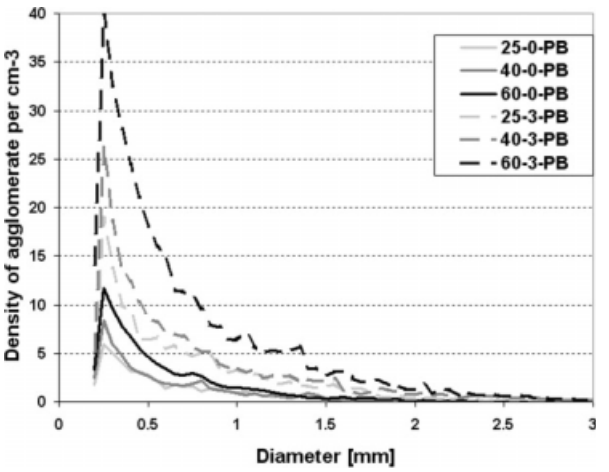

Figure 9 Agglomerates' size distribution. Effect of CMC and dry content during pelletization.

addition of CMC was found to have a major negative effect on the dispersability of the fibers during extrusion. It is noticeable that the dispersability of the cellulose fibers seems to be correlated with the compactness of the pellets. It is believed that the fibers were dispersed during extrusion through the conversion of the shear energy supplied during the compounding. This shear energy was, however, the same for all the pellets since the extrusion process parameter and the fiber content were the same for all the composites. As a result, the difference in fiber dispersion was due to difference in the cellulose fibers pellets only. It is reported in the literature that the poor dispersability of cellulose fibers into polymer matrices is due to the strong fiber-fiber interactions and the incompatibility of hydrophilicity between the cellulose fibers and the polymer matrices.[4,37] By increasing the compactness of the pellets, the contact surface between fibers had increased, promoting more numerous inter fiber interactions. As explained above, the addition of CMC promoted the compactness of the pellets but also created irreversible bonds with and within cellulose fibers, leading to stronger fiber-fiber interactions. Pellets with more inter fiber bonds would therefore need more processing energy to be properly dispersed during extrusion, or they would be less dispersed compared with pellets with fewer fiber-fiber interactions when the same amount of energy was supplied.

The nature of the drying of the cellulose pellets had no effect on the dispersability of the pellets into the PP matrix, see Figure 10. Any difference in agglomerate size or number would probably have been due to larger hornification for the pellets which were oven dried. However, the hornification is not much affected by the drying method but rather by the dryness of the pulp reached.[25] Since all the pellets were oven dried before pelletization to obtained a moisture content below 1 wt %, this oven drying step may have screened any difference which could have existed between the pellets that were air dried or oven dried.

Figure 10 shows the density of agglomerates for 40-0-PB and 40-0-SE. It is clear that 40-0-SE contained more agglomerates than 40-0-PB. The degree of dispersion in 40-0-SE was in the same order as 40-3-PB. The residence time in the extruder was much shorter for the pellets introduced using the side extruder (40-0-SE) than the one introduced by pre-blending (40-0-PB) and therefore resulted in poorer fiber dispersion. This result was in accordance with our previous studies where materials extruded twice showed much better fiber dispersion during extrusion than the material extruded once.[21] Moreover, the measured fiber content was slightly higher for 40-0-SE than for 40-0-PB because of the unstable fiber loading for materials made using pre-blending. It was then expected that 40-0-SE showed more fibers and so more agglomerates than 40-0-PB. It is, however, important to notice that the difference in fiber dispersion was not as much due to the type of feeding technique rather than to the zone of introduction. Therefore, fiber introduced in zone 1 of the extruder using pre-blending with PP matrix or fed using a separate gravimetric feeder would probably show the same degree of dispersion within PP matrix.

Fiber breakage

The average length of the fibers in the composite materials is presented in Table III. The average fiber length was about 0.60 mm for materials without CMC. The moisture content during pelletization had no significant effect on the average length of the

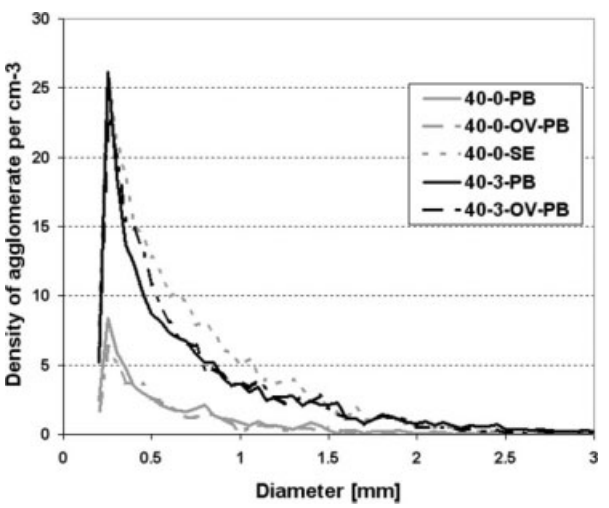

Figure 10 Agglomerates' size distribution. Effect of the drying and feeding methods.

TABLE III
Average Fiber Length of Cellulose Fibers in Pulp, Pellets and Composites

Moisture		Average length [mm] 0% CMC	Average length [mm] 3% CMC	Reduction 0% CMC	Reduction 3% CMC
Paper		1.54[a]			
Fluff pulp		1.54[a]			
Flocks		1.20[a]			
Flocks in composite		0.60[a]		50%[a]	
Pellets	75 wt %	0.82	0.94	47%	39%
	60 wt %	0.86	0.92	44%	40%
	40 w t%	0.78	0.82	49%	47%
Composites	75 wt %	0.62		60%	
	60 wt %	0.57		63%	
	40 wt %	0.57		63%	

[a] CMC and moisture content not relevant.

cellulose fibers. The measurement of the fiber length in composites containing CMC was problematic because the fibers were hardly dispersed after the extraction, keeping the shape of agglomerates. The inter fiber bonds caused by the CMC, which can be irreversible, could be the reason why the fiber could not be dispersed in water. The measured average length of the fibers in composite when CMC was used was however about 0.50mm. This value can only be an indication since the measurement was not representative of the materials, since the agglomerates were discarded by the measurement method.

The feeding zone in the extruder was found to be of importance for the dispersion of the fibers into the matrix. Figure 11 shows the fiber length distribution of 40-0-PB and 40-0-SE composites. There was very little difference between the two materials. The larger shear supplied to the pre-blended fibers was probably not large enough to cause more fiber breakage. However, the fibers could have been damaged without being broken.

Figure 11 presents the fiber length distribution of 40-0-PB and F-0-PB. As visible in the Figure, both fiber length distributions are very close to each others. Despite the longer fibers of the flocks at the introduction in the extruder, the fibers in the composites had similar length. The fiber breakage during extrusion was therefore more severe for flocks than for pellets. The efforts in limiting fiber breakage during pelletization were therefore negligible compared with the fiber breakage caused by extrusion.

CONCLUSIONS

Effects of moisture and CMC contents on the pellets properties:

Table IV summarizes the effects of carboxymethyl cellulose (CMC) and moisture content during pelletization on the properties of the pellets and the composites. Cellulose fibers were successfully pelletized when the moisture content of the pulp was between 40 and 75 wt %. CMC was found to be a good processing aid for the pelletization of cellulose fibers and to slightly limit fiber length reduction during pelletization. Moisture content had no significant effect on fiber breakage during processing.

The dispersability of the fibers during extrusion was in inversely related to the compactness of the pellets. Increased compactness of the pellets was caused by low moisture content during pelletization and use of CMC. CMC was creating additional inter fiber bonds. However, the compactness pellets limited the dispersability of the fibers in the extruded product.

Evaluation of the process parameters for compounding cellulose-PP composites:

Introduction of compact pellets using the side extruder was not possible because the equipment was

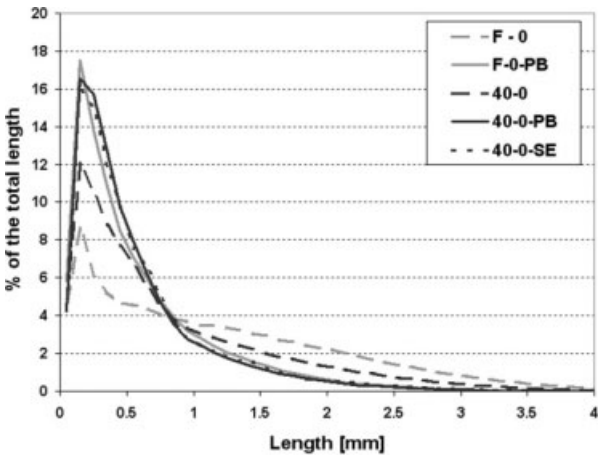

Figure 11 Fiber length distribution in composite materials.

TABLE IV
Effect of the Preparation Parameters on the Pellets Properties

Properties	Parameters		
	High dry content during pelletization	Addition of CMC prior pelletization	Oven drying
Processing of pelletization	Best about 40 wt %	+ + +	n.a.
Maintain fiber length during pelletization	0	+	0
Apparent density	+ +	+	0
Pellet dimensions	+	+ +	0
Pellet compactness	+	+ +	0
Dispersability during extrusion	−	− − −	0
Fiber breakage during extrusion	0	n.a.	0

+ positive effect, − negative effect, 0 no effect, n.a. non applicable.

adapted to neither the dimensions nor the compactness of the pellets. Introduction of pellets using side extruder was, however, stable and controllable whereas segregation occurred when the materials were pre-blended. Early introduction of the fibers in the extruder was beneficial for the fiber dispersion and did not cause significant additional fiber length reduction.

The efforts in limiting fiber breakage during pelletization were useless because they were balanced by larger breakage during extrusion. The final extruded composites had about the same fiber length, regardless of the process. The drying method had no effect on the pellets properties.

Best cellulose fiber pulp for extrusion application was therefore the pulp with a moisture content of 75 wt % and no CMC. The presence of CMC made cellulose pellets stiffer, which could be beneficial for transport, storage and handling, but which is clearly a drawback when the fibers have to be dispersed in subsequent processing like extrusion.

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