BACTERIAL CELLULOSE REINFORCED THERMOPLASTIC COMPOSITES: PRELIMINARY EVALUATION OF FABRICATION AND PERFORMANCE

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Mechanical properties of polyethylene (PE) composites were evaluated as a function of the addition of bacterial cellulose (BC). It was found that BC could improve the mechanical properties of the composites with or without the combination of traditional wood fiber. The improvements were affected by post-treatment. It was confirmed that BC had a significant influence on impact strength. The pellicle form of BC was able to achieve superior impact strength compared to the fluffy form of BC, but had similar effects on the tensile strength in comparison to the composites with fluffy BC.

Keywords: Bacterial cellulose; Nanofiber; Polyethylene, Wood fiber; Wood composites

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INTRODUCTION

Polyethylene is a petroleum-based synthetic polymer that is difficult to biodegrade. By contrast, cellulose is the most abundant biopolymer on earth, as the major component of plant cell walls, and it is also a representative of microbial extracellular polymers. Cellulose has drawn much attention for being biodegradable and renewable (Sanjeev and Mohanty 2007; Wu 2009), inexpensive and capable of being produced in large quantities (Pritchard 2007; Mathur 2006), for its low disposal cost (Selke and Wichman 2004; Wu 2009), and for the fact that wood composites are bioproducts, at least in part (Satyanarayana et al. 2009; John and Thomas 2008). Some bacteria produce cellulose (called biocellulose or bacterial cellulose, BC) (Brown 1978; Watanabe et al. 2004; Morán et al. 2008). Wood fiber and BC have identical chemical structure, but different physical and chemical properties (Chau et al. 2008; Kumar et al. 2007; Duhardt et al. 2005). Nanofibers or microfibrillated/nanofibrillated cellulose are principally produced by mechanical disintegration of wood-derived fiber bundles into submicron nanofibril bundles (10-50nm in diameter) with the help of enzymatic pre-treatment (Klemm et al. 2009; Ankerfors and Lindström 2007; Ankerfors and Lindström 2009), which is applied before mechanical procedures in order to ease the delamination and to decrease the energy consumption. But nano-scaled BC is mainly produced by an acetic acid-producing bacterium, *Gluconacetobacter xylinum*, taking advantage of the isolation of a particularly efficient strain (Yamasa et al. 1997; Yamada 2000; Chao et al. 1997).

BC has some characteristic physical and chemical properties such as high crystallinity and hydrophilicity, ultrafine network architecture, purity (free of lignin and hemicelluloses) (Goelzer et al. 2009), and moldability during formation (Kumar et al. 2007; Udhardt et al. 2005). The unique properties may result from an ultrafine reticulated structure from the nascent chains of BC with a fiber-width of approximately 50 nm. BC has been used in enzyme immobilization (Wu and Lia 2008; Jonas and Farah 1998), and biomedical applications (Wan et al. 2006; Kim et al. 2010), whereas it has been less considered for papermaking (Surma-Ślusarska et al. 2008; Basta and El-Saied 2009), or for food industry applications (Okiyama et al. 1993), due to its higher cost relative to commercial cellulosic pulps. BC has a high potential for the development of a new class of truly green composites as a reinforcement agent due to its high Young's modulus, up to 134GPa (Klemm et al. 2006), and the fact that is derived from renewable resources. The incompatibility problem between a hydrophobic matrix and hydrophilic fibers applies to the hydrophilic BC, not just to the types of cellulosic fibers typically used in such application. It follows that interfacial compatibility should be improved by use of a coupling agent (Keener et al. 2004; Mohanty et al. 2006; Li and Matuana 2003; Gu et al. 2009a) as well as an initiator (Gu et al. 2009a; Krupa and Luyt 2001; Gaboyard et al. 2002; Nogellova et al. 1998), resulting in the formation of chemical bonding between the wood fiber and the matrix (Carlborn and Matuana 2006; Lu et al. 2000).

In this study, bacterial cellulose composites were fabricated to study the reinforcement behavior of BC on the mechanical properties. The effect of BC reinforcement was also investigated with the addition of chemi-thermo-mechanical pulp (CTMP) fiber or not. In addition, a post-treatment method of BC was also evaluated based on the studies of the reinforcement of BC in different formation.

EXPERIMENTAL

Materials and Methods

Thermoplastic

Linear low density polyethylene (PE, Novacor® HI-0753-H) was donated by NOVA Chemicals. Its melting mass-flow rate is 1.0g/10min. Its specify density is 0.92g/cm^3.

Wood fiber

Industrial CTMP of yellow birch (*Betula alleghaniensis*) was employed. The wood pulp fibers were air-dried and ground with a grinder (CWB Instruments Inc.) at our laboratory to produce fine particles. Particles that passed through a 20-mesh but were retained on a 60-mesh screen were employed in this study.

Coupling agent

Maleated polyethylene (G2010) was supplied by Eastman chemical company (Kingsport Tenn.). Its maleic acid graft content is 1.5wt%, and the molecular weight is 15,000.

Initiator

Dicumyl peroxide (98% active DCP) supplied by Sigma Chemical Co. was used as an initiator. Its halftime is 1 min at 171ºC.

Scanning electron microscope (SEM) investigation

SEM investigations of BC were conducted as described by Hornung et al. (2007). Sample stubs with fibers were surface-metalized by sputter coating (Polaron-Model SC 7620) with evaporated Au metal (4 nm in thickness) after it was mounted on an aluminum stub, and then was carried out by SEM (JSM 5500, Jeol, Japan) at an acceleration voltage of 15kV at the Integrated Pulp and Paper Center, University of Quebec at Trois-Rivieres.

Preparation of Bacterial Cellulose

Bacterial cellulose materials were produced by *Gluconacetobacter xylinum* AX 5 (culture collection fzmb GmbH-Research Center of Medical Technology and Biotechnology, Geranienweg 7, 99947 Bad Langensalza, Germany) in classical Schramm/Hestrin (SH)-medium (Schramm and Hestrin 1954), which contains glucose as described in previous reports (Hornung et al. 2006a,b, 2007). BC sheets were produced by static fermentation, washed with water after fermentation, and cut as shown in Fig. 1.

Fig. 1. Picture of original BC made by static fermentation (the source: fzmb GmbH)

In order to study the effect of a post-treatment method on the mechanical properties of PE composites as well as the assistance of wood fiber, the original BC was mechanically treated by different post-treatment methods after it was harvested. Two different physical forms, were obtained, fluffy and pellicle forms as shown in Fig. 2, depending on the processing conditions. The fluffy sample 1 (B1) was obtained by use of a cutting mill with the wet material and milling after freeze drying, using a centrifugal mill. Pellicle sample 2 (B2) was first freeze-dried in its original wet form, and then milled by a centrifugal mill. The particle sizes of B1 and B2 were not examined in our work. However, the fibers were similar in diameter (50 to 100nm), as shown in Fig. 3. Still, BC

Fig. 2. Pictures of fluffy (left, B1) and pellicle (right, B2) BC after treatments

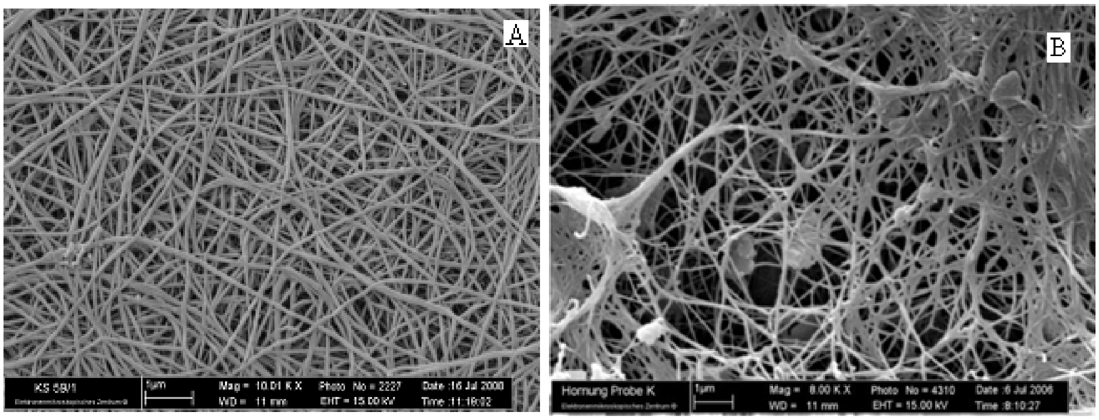

Fig. 3. SEM of the general structure of original BC (the source: fzmb GmbH)

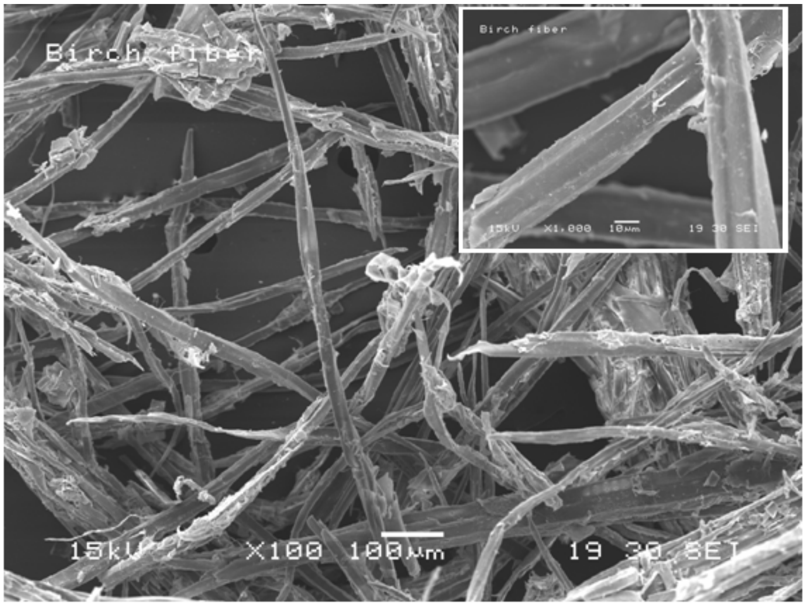

Fig. 4. SEM of CTMP birch fiber

was different from wood-derived nanofibrillated cellulose. BC has denser networks than wood fiber, as indicated in Fig. 3A, and also has a highly branched, three dimensional, and reticulated structure, as shown in Fig. 3B. By contrast, wood fiber is un-branched, as shown in Fig. 4.

Preparation of PE Composites

Compounding

All of the sheets were prepared using a two-roll mill, Thermon C.W. Brabender (Model T-303), according to the conditions as follows to study the influence of wood fiber with the presence of the compatibilizers as well as the bacterial cellulose on the mechanical properties, as formulated in Table 1. The amounts of maleated PE and initiator were selected according to our previous study (Gu et al. 2009a). Twenty weight per cent PE with G2010 was melted on rollers at 170°C. The residual (80wt %) PE with or without bacterial cellulose was added and blended for 3 min at 60 rpm, and then wood fibers were added or not, with continued blending for 4 min (the total time is 7 min). The mixture was peeled from the roller and re-blended 5 times, each time for 3 min, to obtain a uniform composite sheet. Finally, the sheet was removed from the roller and made into strips with a knife according to the molder size after DCP had been added and blended for 3 min.

Table 1. Composition of the Composites

Code	Composition, wt%					
	PE	Wood fiber	B1	B2	G2010	DCP
A	100	--	--	--	--	--
B	96.8	--	--	--	3	0.2
C1	95.8	--	1	--	3	0.2
C2	95.8	--	--	1	3	0.2
D	70	30	--	--	--	--
E	66.8	30	--	--	3	0.2
F1	65.8	30	1	--	3	0.2
F2	65.8	30	--	1	3	0.2

Molding

22 specimens (10 for tensile strength testing, and 12 for tensile impact strength testing) for each sample were simultaneously obtained by pressing the sheet trips into a dog-bone shaped mould (ASTM D638 Type V for tensile strength and ISO 8256 Type II for tensile impact strength). The approximate dimension of a tensile specimen was 0.30-0.32cm in width and 0.27-0.33cm in thickness, while the width of impact specimen was 0.29-0.36cm and the thickness was 0.17-0.18cm. The specifications for the use of the two-roll mill were: 30 cm length, 15 cm radius, 0.6 gear ratio, and 60 rpm roll speed.

Mechanical Tests

All of the specimens were conditioned at 23^{o}C and 45% relative humidity, and then width and thickness were measured after polishing. The tensile test was performed using an Instron machine (Model 4201) according to ASTM D638, while tensile impact strength testing was carried out by means of a Zwick tester following the DIN 53448 method under 23°C and 45% relative humidity.

RESULTS AND DISCUSSION

The characteristics of B1 and B2 were strikingly different; B1 could be described as a fluffy form and B2 could be described as a pellicle form. This difference was attributed to the manner of physical preparation of each type of sample. However, their chemical functions should be the same when BC is well dispersed in the composites. In fact, it is difficult to achieve a uniform distribution during a melting and compounding process. Nonuniformities of the distribution of BC can be discernible due to its fibril-like aggregations, in particular for hydrophobic systems, even in cellulose acetate butyrate composites (Gindl and Keckes 2004). Obviously, the planar B2 was dispersed more easily than 3-D conformation of B1 when BC chains were exposed to high driving shear force during compounding, because there were more large fibril-like spots of B1 compared to B2 according to our observations.

In the following discussion, the mechanical properties of the composites are compared after the addition of wood fiber and BC in the presence of a coupling agent and initiator, as depicted in Table 2. As in our previous report (Gu and Kokta 2010), the coupling agent and initiator could deteriorate the tensile strength, increase the elongation, and lower their tensile modulus (Sample B). But it is worth noting that it could improve the impact strength by 23% from 45.4kJ/m² for neat PE up to 55.8kJ/m² for compatibilized PE. There was an indication that a small amount of BC improved the interaction between the wood fibers and the synthetic polymer matrix due to its branched structure, which enhanced tensile strength by 5 to 7% compared to coupled PE (Sample B), that is neat PE functionalized by coupling agent and initiator. Unlike tensile strength, the impact strength of compatibilized PE decreased as a small amount of BC was introduced, as shown in Table 2. Neither B1 nor B2 addition could achieve superior impact strength compared to virgin PE (Sample A) or coupled PE (Sample B) due to their fibril-like aggregations from their denser networks and their reticulated structure. It is important that the bacterial cellulose composite reinforced with B2 (Sample C2 44.9kJ/m²) had relatively high impact strength in contrast to B1 (Sample C1 26.4kJ/m²), resulting from its relatively uniform dispersion as a consequence of its physical form.

Because BC use tends to result in a high tensile modulus (Klemm et al. 2006), the composites reinforced with 1wt% BC had high tensile modulus, which was increased by 20-30% compared to neat PE, and 50-60% compared to compatibilized PE without any fiber added (either wood fiber or BC-fiber). Obviously, the improvement of elongation was significant from 11% of the coupled PE down to 8-8.4% as 1wt% BC was employed. It seems that both pellicle and fluffy BC yielded similar improvements in tensile strength, tensile modulus, and elongation. It follows that the contributions come from the ester bonds formed between the hydroxyl groups of fiber and the anhydride groups of the coupling agent (Carlborn and Matuana 2006), not being related to physical morphology, if they are in the same scale as well as distributed well. The direct evidence of chemical bonds between bacterial cellulose and the matrix has not been proved, but the interfacial adhesion among the composites was surely improved due to BC making up the wood fiber structures (Juntaro et al. 2008).

In fact, the two post-treatment methods of BC showed no influence on their chemical composition due to their being generated by same bacteria, but having contrasting morphologies. The influence of morphology alone was also evaluated indirectly, due to that fact that the chemical composition of BC is being similar to that of wood fiber. Whatever the explanation, in the absence of traditional wood fiber, with a small amount of BC addition, it could be difficult to obtain superior strength higher than that of neat PE due to the fact that excess maleated polymer could form a thick attaching hydrophobic layer, leading to slippages within the hydrophobic matrix. Considering the high manufacturing cost of BC, an amount of wood fiber was introduced to investigate the changes of the mechanical properties of bacterial cellulose composites.

It was observed that a composite with 30wt% wood fiber added had the weakest tensile strength and impact strength (Sample D 18.1MPa and 12.5kJ/m²), respectively, due to poor adhesion. It is also well known that coupling agent and initiator could improve both impact strength and tensile strength of the composite (Sample E with 34.6MPa and 40.9kJ/m², respectively) when wood fiber is added with the formation of ester linkages (Carlborn and Matuana 2006; Lu et al. 2000).

Table 2. Mechanical Properties of the Composites

Code	Mechanical properties (Average value)			
	Impact strength kJ/m²	Tensile strength[a] MPa	Elongation[b] %	Tensile modulus MPa
A	45.4(4.4)[c]	25.4(1.9)	10.4(1.4)	771(90)
B	55.8(5.8)	19.7(1.2)	11(0.7)	644(83)
C1	26.4(2.2)	20.6(1.3)	8(0.8)	962(68)
C2	44.9(4.4)	21.1(0.8)	8.4(0.9)	1011(87)
D	12.5(2.9)	18.1(1.2)	2.5(0.3)	1188(108)
E	40.9(3.9)	34.6(1.3)	9.1(0.7)	900(85)
F1	42.3(1.8)	36.9(1.3)	9.3(0.7)	951(101)
F2	54.3(1.9)	36.8(0.8)	9.5(0.7)	893(44)

Notes: a: tensile strength means stress at the max load; b: elongation means strain at max load; c: the data in the brackets represent standard deviation

Due to the fact that BC could attach in situ to wood fiber via strong adhesion caused by the high self-affinity of cellulose through hydrogen bonding (Ross et al. 1991) as well as their similar polarities, the introduction of BC in small amounts could increase the impact and tensile strength of the composites (Sample F1 and F2) compared to the composite without BC-fiber present (Sample E). Moreover, with the assistance of wood fiber, BC not only improved the tensile strength of PE-wood composites (Sample F1 and F2), but also enhanced their impact strength, which differed in its behaviors in PE composites employed with BC only (Sample C1 and C2). Pommer et al. (2008) and Juntaro et al. (2008) demonstrated the attachments of BC on the surface of sisal fiber in water-based culture medium. Juntaro et al. (2008) reported that the interfacial adhesion between PLA and the sisal fiber was enhanced when BC attached onto fiber surfaces, implying a relatively high apparent interfacial adhesion (Juntaro et al. 2008). Although this attachment occurred in culturing, it shows us that there exists a way for BC to deposit on wood fiber surfaces, even by compounding, and at least it could disperse better than those without the presence of wood fiber. Still, their similar polarities also contribute to BC achieving more uniform dispersion accompanying the distribution of wood fiber, in particular under strong compressive and shear forces. Although a low concentration of BC is given, the dominant effect is almost certainly related to an improvement in the apparent interfacial shear strength, which allows the primary fibers, that is wood fiber, to contribute more effectively to the overall composites performance. So, both impact and tensile strength were improved after small amounts of B1 and B2 were included in the composites F1 and F2, which also contributed with a little high strain as indicated in Table 2.

The difference of impact and tensile strength of F1 and F2 came from their physical forms. It is an interesting finding that B1 raised the impact strength by 60% from 26.4kJ/m² for the bacterial cellulose composite (C1) up to 42.3 kJ/m² for the composite with 30wt% birch fiber present (Sample F1). As a flipside to the improvement of the dispersion of B1 in the composite, B2 enhanced impact strength by 21% from 44.9kJ/m² of C2 up to 54.3kJ/m² for F2. Even though pellicle BC (B1) would be dispersed better than fluffy BC (B2), it was still found that the distribution of fluffy BC (B1) was improved significantly with the help of traditional wood fiber compared to pellicle BC (B2), with the consequence that impact strength was greatly improved. Benefiting from this contribution, the tensile strength was also improved by feeding a small amount of BC in addition. It has been reported that the interfacial adhesion between PLA and the sisal fiber was enhanced when BC was attached onto fiber surfaces, implying a relatively high apparent interfacial adhesion (Juntaro et al. 2008). However, some difference with respect to the contribution to tensile strength of the composites with the reinforcement of BC helping with additional dispersion via wood fiber was observed, where B1 addition could achieve 79% enhancement from 20.6MPa of C1 up to 36.9MPa of F1, while with B2 addition the adhesion was increased 75% from the value at 21.1MPa of the composite without wood fiber (Sample C2) up to the value at 36.8MPa of the composites with wood fiber (Sample F2). So, it is concluded that wood fiber could help BC achieve a more uniform dispersion. The improved distribution of BC will optimize the distribution of the networks of coupling agent and cellulose chains (wood fiber and BC), which results in superior interfacial adhesion to achieve enhanced impact and tensile strength.

In case of the biocomposites including BC and wood fiber, more fibers introduced could induce the molecules of maleated polymer to covered the fiber surface with a thin hydrophobic layer, leading to embedding them into the PE matrix and preventing slippages from occurring (Keener et al. 2004; Mohanty et al. 2006; Gu et al. 2009b). Although a very small amount of BC was introduced, as long as the particles were embedded and well combined with traditional wood fiber, it was possible to increase the impact strength of the composites by 3% and 33% through reinforcement with B1 and B2 compared to the composites without BC (Sample E), respectively. In contrast to impact strength, the tensile strength of F1 had nearly the same change as F2 after a small amount of BC was added, where both of them were increased in the range of 6% compared to wood composites without the presence of BC (Sample E). B1 and B2 had different effects on impact strength, but less on tensile strength, elongation, and modulus due to their different reinforcement mechanisms. The contribution of impact strength came from the formation of networks in the wood composites. Obviously, a more uniform distribution of pellicle BC (B2) could produce more networks than fluffy BC (B1), leading to improved impact resistance, increased elongation, and decreased tensile modulus, even though such effects were slight. But for tensile strength, the contribution mainly depended on the amount of bonds formed, which was limited by the amount of exposed BC.

Decreased elongation and increased tensile modulus of composites that had not been treated with either a coupling agent (G2010) or a crosslinking agent (DCP) were discovered in this work; this is attributed to a high tensile modulus of wood fiber acting as backbones in the matrix (Keener et al. 2004; Yuan et al. 2008) as well as a remarkably high modulus of elasticity of BC due to its super-molecular structure (Yamanaka et al. 1989). The elongation difference is attributed to improved interfacial adhesion. Improved compatibility in some cases could allow coated wood fibers to flow past one another within a very small range, which could explain a slight increase of elongation. Such elongation behavior is typical for reinforced thermoplastic in general and has been reported by many researchers (Raj et al. 1989; Bledzki and Gassan 1999). However, the macrophysical tensile modulus is still dominated by the contributions of the backbone fibers, such as high tensile modulus of rupture, which is little affected by the addition of BC, due to its small amount.

CONCLUSIONS

Though the impact strength of neat PE could be improved by the addition of coupling agent and initiator (55.8 kJ/m²), no further improvement in impact strength was achieved by the addition of wood fiber and BC (54.3 kJ/m²), as shown in Table 2. However, by employing both wood fiber and bacterial cellulose, the resulting composites could increase in modulus while maintaining or even increasing their tensile strength. Moreover, it was also possible to optimize its impact strength.

A small amount of bacterial cellulose (BC) introduced into the composites, in combination with addition of traditional wood fiber, was able to improve impact strength, tensile strength, and their elongation. The post-treatment of BC had a great impact on impact strength, both in the presence and absence of wood fiber. Impact strength was much more sensitive to the distribution of celluloses compared to tensile strength. It seems that BC without mill cutting yielded greater improvement, so that pellicle BC gave more favorable results than fluffy BC. But the difference could be reduced with the assistance of wood fiber in a good dispersion of BC. In addition, the elongation results showed similar trends to tensile strength. However, tensile modulus originates from the contributions of the backbone of wood fiber reinforcement, leading to a reduced influence of the type of BC. This improvement will contribute a high deformation resistance of the resulting composite. It is concluded that pellicle BC can provide superior improvements to mechanical properties of a reinforced plastic composite.

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