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Properties of Natural Rubber Latex Filled with Bacterial Cellulose Produced from Pineapple Peels

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Abstract

Bacterial cellulose (BC) is natural biopolymers with unique properties such as high purity and high tensile strength. These properties considerably support for use as filler in rubber applications. In this research, BC was produced from fermentation by *Acetobacter xylinum*. Additionally, pineapple peels (agricultural waste) were evaluated for carbon sources in BC's production process. Natural rubber latex (NRL) compound was then incorporated BC with various loadings of 0, 5, 10 and 15 phr (parts per hundred of rubber) for enhancement properties of NRL. Furthermore, NRL filled with BC (NRL-BC) compounds were prevulcanized at 70°C for 2 h and then NRL-BC sheets were finally prepared. NRL-BC sheets were examined in the term of tensile properties, tear strength, and dynamic mechanical properties (DMA). From the results, with incorporation of BC, it was observed that not only tensile strength, tensile modulus and tear strength slightly increased but also DMA result clearly showed increase in storage modulus. Moreover, results revealed that NRL containing 10 phr of BC provided optimum properties.

Keywords: Natural rubber latex; Bacterial cellulose; Pineapple peel; Filler

Introduction

Although natural rubber (NR) is known to exhibit numerous outstanding properties, reinforcing fillers are necessarily added into NR in some cases in order to gain the appropriate properties for the specific applications. A wide variety of fillers are used in the rubber industry for various purposes of which the most important are reinforcement and reduction in material costs, and improvements in processing. Carbon black and silica is widely used as reinforcing filler to improve physical and mechanical properties of rubber(1,2). Additionally, the use of fibers is another successful method for enhancing properties of NR. Some examples of fibers used to reinforce NR are aramide, nylon and cellulose(3-5).

However, the sustained demand for environment-friendly reinforcing filler still is an interesting and challenging for rubber industry. Then, the use of some other fillers from the natural resources and by-product as alternative fillers in NR has been carried out(6-8). Interestingly, cellulose produced by bacteria namely *Acetobacter xylinum* called bacterial cellulose (BC) has been used as reinforcing agents for natural and synthetic polymers(9). Bacterial Cellulose or BC is natural biopolymers which composed of high purity of cellulose without any impurities such as lignin, pectin, and hemicellulose(10). Generally, glucose has been used as a carbon source for BC biosynthesis by *A. xylinum*. It has been reported, however, that BC was also synthesized from other carbon sources, such as 5- or 6-carbon monosaccharides(11). In this BC preparation, pineapple peels (agricultural waste) were evaluated for carbon sources in BC production process. Then, a feasibility study was carried out on the utilization of BC produced from pineapple peels as a filler in natural rubber latex (NRL). The effects of BC loadings on mechanical of NRL vulcanizates were systematically studied for further application.

Material and Experimental Procedures

Preparation and characterization of BC

For preparation of bacterial cellulose sheets, pineapple peels juice was firstly prepared and was

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further used for carbon source in BC synthesis process. Bacterial cellulose sheets were obtained from fermentation of the bacterial namely *Acetobacter xylinum* (TISTR 975) in medium solution composing pineapple peels juice, ammonium sulfate ($(NH_4)_2SO_4$), sucrose ($C_{12}H_{22}O_{11}$) and acetic acid (CH_3COOH). The mixtures were incubated for 1 week at 28°C in beaker 1000 mL. After 7 days of incubation, BC sheet (1.5 mm of thickness) was finally obtained. These sheets were washed in 0.5 M NaOH at 70°C in order to remove bacteria and then several times in water until neutral pH. BC sheets were characterized by Fourier Transform Infrared Spectrophotometer (FTIR), thermogravimetric analyzer (TGA) and scanning eletron micrpscope (SEM).

Preparation of NRL/BC composites

The loading of BC filled NRL composites was varied from 0 to 15 phr. BC was incorporated to NRL and chemical agents as shown in Table 1. The mixture was then mixed using digital magnetic stirrer for 2 h at a setting temperature of 70°C. The NRL filled with BC sheets were obtained by pouring prepared the mixture of NR with BC in to plate and vulcanizing at 100°C for 1 h.

Table 1: Compounding formulation for natural rubber composites.

Ingredient (phr)	Formulations 0	5	10	15
Natural rubber latex	100.0	100.0	100.0	100.0
Bacterial Cellulose	0.0	5.0	10.0	15.0
Dispersion Sulfur	5.0	5.0	5.0	5.0
Dispersion ZDEC	3.0	3.0	3.0	3.0
Dispersion CPL	2.0	2.0	2.0	2.0
Dispersion Zinc Oxide	3.0	3.0	3.0	3.0

Measurement of tensile and tear properties of NRL/BC composites

NRL/BC sheets having a thickness of about 2 mm were used for tensile and tear testing according to ASTM D412 and D642, respectively. The measurements were carried out using an universal testing machine (Instron Model 3366) with a crosshead speed of 500 mm/min. At least, 5 specimens were tested for these properties and the average values were reported.

Measurement of dynamic mechanical analysis of NRL/BC composites

Dynamic mechanical analysis (DMA) measurement was carried out with dynamic mechanical analyzer (Gabo ExplexorTM 25N) working in the tension mode. Tests were performed on temperature varied from -80 to 20°C.

Results and Discussion

Characterization of BC

Determination of some characteristics of BC used in this study was carried out. FTIR spectra of BC as depicted in Figure 1 shows the broad peaks at 3,344 cm^{-1} indicating the presence O-H stretching of hydroxyl group. The absorption peak at 2932 cm^{-1} and the region 1300-1400 cm^{-1} is attributed to CH and CH_2 groups. The present peak in at 1,107 cm^{-1} is attributed to C-O-C strethching.[12]

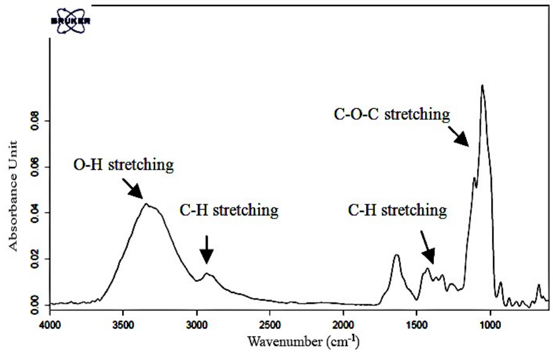

Figure 1. FTIR spectra of bacterial cellulose (BC).

The thermal degradation of BC are illustrared in the term of percentage of weight loss with temperature as shown in Figure 2. From the TGA thermogram of BC, on the first step of weight loss, the slightly weight loss of BC at temperature below 200°C can be observed. This is due to the moisture evaporated. Furthermore, BC shows degradation temperature around 350°C. Additional, SEM micrographs are demonstrated in Figure 3.

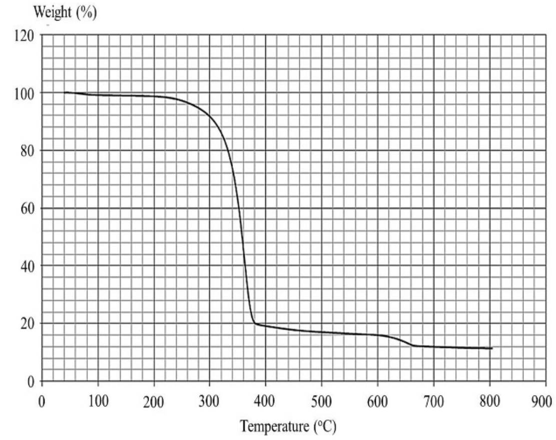

Figure 2. TGA thermogram of BC.

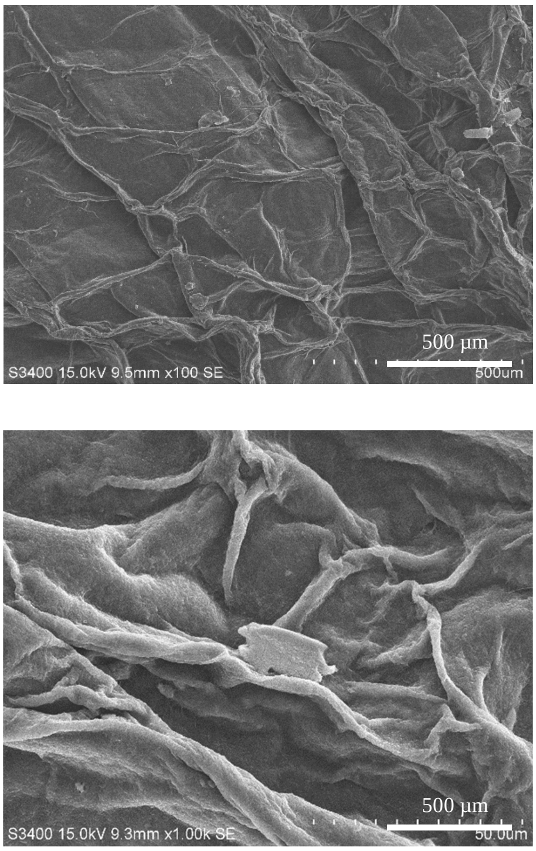

Figure 3. SEM micrographs of BC sheets.

Tensile and tear properties of NRL filled with BC composites

Tensile strength, elongation at break and modulus at 100% elongation (M100) are shown in Figures 4-6, respectively. From the result, it can be seen that the tensile strength of BC filled NRL in Figure 4 slightly improves with incorporation up to 10 phr of BC. The decrease in tensile strength is observed when BC was added after 10 phr. This is probably due to the poor dispersion of BC and the tendency of the BC to form cluster. The elongation at break of BC filled NRL is illustrated in Figure 5. It can be noticed that the elongation at break of NRL/BC composites decreases with increasing BC loading. The deceasing trend of elongation at break is attributed to increase in stiffness[13,14]. As expected, the enhancement in M100 as shown in Figure 6 because the increase in stiffness of NRL vulcanizates. Interestingly, it was reported that BC existed in crystalline phase resulting in enhancing modulus value[9]. Figure 7 displays the tear strength of BC filled NRL composites. The result shows that tear strength gradually increases with incorporation of BC up to 10 phr. It can be explained that the poor dispersion of BC in NRL as well as the result of tensile strength as discussed previously.

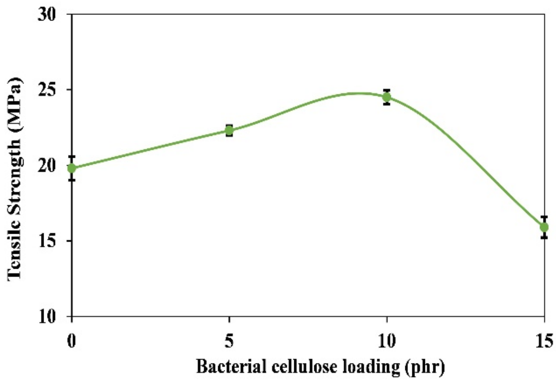

Figure 4. Tensile strength of NRL filled with BC composites.

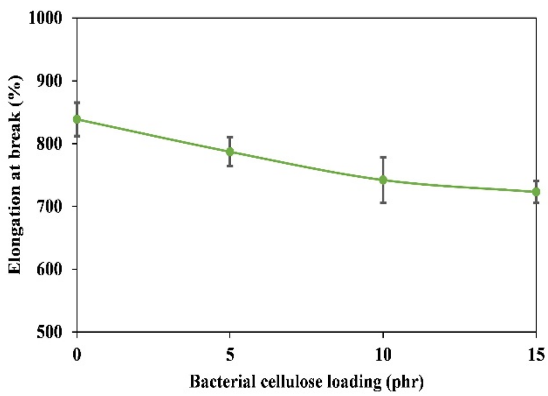

Figure 5 Elongation at break of NRL filled with BC composites.

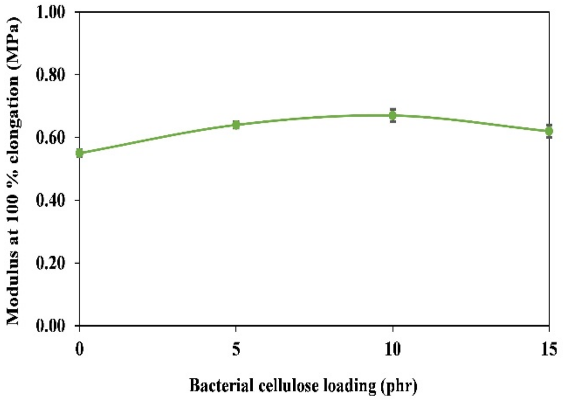

Figure 6. Modulus at 100% elongation (M100) of NRL filled with BC composites.

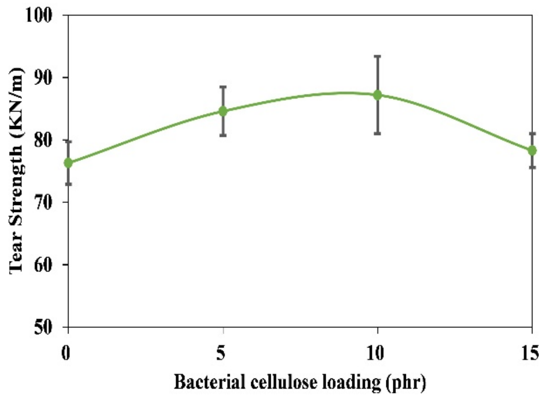

Figure 7. Tear strength of NRL filled with BC composites.

Dynamic mechanical analysis of NRL filled with BC composites

DMA results which are the plot of storage modulus of NRL/BC composite as a function of temperature as shown in Figure 8. Normally, storage modulus is represented elastic of material. From the results, NRL shows glassy stage at temperature below glass transition temperature (T_g) and drop in its storage modulus around the temperature corresponding to the glass-rubber transition, and then remaining constant with increasing temperature.[15] The storage modulus of the composites slightly increase with increasing BC loading, especially in the glassy stage. It can be noticed that the 10 phr of BC in NRL shows highest storage modulus. However, the modulus of the composites at the rubbery plateau are no different change after incorporation BC.

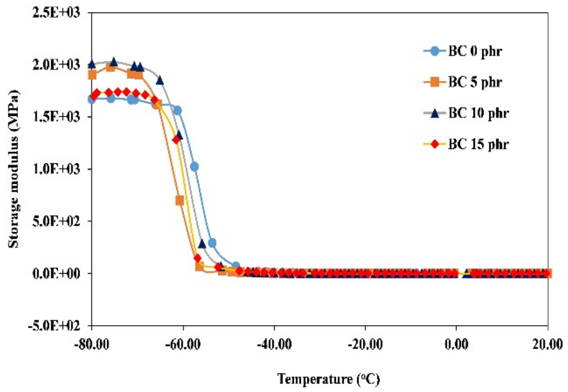

Figure 8. Storage modulus of NRL filled with BC composites.

Conclusions

In this studty, BC was produced from fermentation by *Acetobacter xylinum* using pineapple peels for preparation carbon sources in BC's production process. Obtained BC were characterized by FTIR, TGA and SEM. Furthurmore, natural rubber latex (NRL) was then incorporated BC as mention eariler with various loadings of 0, 5, 10 and 15 phr (parts per hundred of rubber). NRL filled with BC composites were examined in the term of, tensile properties, tear strength, and DMA. From the results, with incorporation of BC, it was observed that not only tensile and tear properties slightly increased but also DMA result clearly showed increase in storage modulus. Moreover, results revealed that NRL containing 10 phr of BC provided optimum properties.

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