PERFORMANCE EVALUATION OF BACTERIAL CELLULOSE REINFORCED POLYETHYLENE COMPOSITES

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ABSTRACT

PERFORMANCE EVALUATION OF BACTERIAL CELLULOSE REINFORCED POLYETHYLENE COMPOSITES. This paper discusses the influence of bacterial cellulose reinforced polyethylene composites on the physical and mechanical properties. Composites consisting of polyethylene and bacterial cellulose were prepared by extrusion and compression process. The content of bacterial cellulose subjected to polyethylene was set from 0-50 (w/w)% with the addition of 2 % Polyethylene Maleic Anhydride (MAPE). The effect of bacterial cellulose content in the composites was evaluated. The tensile strength and elongation at break of composite declined with increasing of bacterial cellulose, but the Young's modulus increased, demonstrating that composite becoming more rigid. At 23 $^{\circ}$C 50 % RH, tensile strength, break strain and modulus of elasticity of polyethylene was 21.2 MPa, of 270 % and 763 MPa respectively. With the bacterial cellulose loading up to 50 %, tensile strength dropped to be half, break strain to be less than 0.5 %, but Young's modulus increased more than 230 %. In addition, the composite became more hydrophilic and more amorphous loading with bacterial cellulose.

Keywords : Bacterial cellulose, Polyethylene, Composites, Mechanical properties

ABSTRAK

EVALUASI KINERJA KOMPOSIT POLIETILEN YANG DIBERI SELULOSA BAKTERIA SEBAGAI PENGUAT. Makalah ini membahas pengaruh penambahan selulosa bakteria sebagai penguat terhadap sifat fisis dan mekanis komposit polietilen. Komposit dibuat dari campuran polietilen dan selulosa bakteria melalui proses ekstruksi dan kompresi. Kandungan selulosa bakteria terhadap polietilen dibuat 0 % hingga 50 % (b/b) disertai penambahan 2 % *Polyethylene Maleic Anhydride* (*MAPE*). Pengaruh kandungan selulosa bakteria dalam komposit telah dikaji. Nilai kuat tarik dan kemuluran dari komposit menurun dengan penambahan selulosa bakteria, tetapi modulus Young meningkat, menunjukkan bahwa dengan komposit semakin kaku. Kuat tarik, kemuluran dan modulus Young pada kondisi suhu 23 $^{\circ}$C dan kelembaban 50 % masing-masing adalah 21,2 MPa, 270 % dan 763 MPa. Dengan penambahan selulosa bakteria sampai 50 % nilai kuat tarik menurun menjadi setengahnya, kemuluran menjadi kurang dari 0,5 % tetapi modulus Young meningkat lebih dari 230 %. Selain itu, komposit menjadi lebih bersifat hidrofilik dan lebih amorf dengan penambahan selulosa bakteria .

Kata kunci : Selulosa bakteria, Polietilen, Komposit, Sifat mekanik

INTRODUCTION

Polymeric materials are present in all kinds of applications, due to its ease of processing and productivity, excellent properties light-weight and low cost. The light weights of these composites also increase the energy efficiency for machinery and transportation. In most of these polymer composite applications, the properties of polymers are modified using fillers and fibers to suit the high strength/high modulus requirements. One of the main categories of polymer composites is represented by polymers filled with synthetic filler or reinforcement such as glass fibers, which fit to a number of applications. The use of glass fiber resulted in the high modulus, high strength, and good moisture resistance composites [1], but the density of glass fiber used for composites is 2.6 g/cm^3 higher than natural fiber such as flax fibers of 1.5 g/cm^3.

In addition, in application and production areas of the factory where glass fiber components are applied, skin irritations and respiratory diseases caused by the inhalation of fiber dust may happens to the workers. In this regards, the use of natural fibers as fillers or reinforcement for both thermoplastics and thermosetting polymer composites offers an interesting alternative replacing petroleum based resources, due to their low cost, low density, widely available, non abrasive, sustainable and biodegradable [2]. Natural fiber used

can be originated either from agricultural activity or forestry resources, which are also referred to as cellulosic or lignocellulosic fibers related with the main chemical component, namely cellulose and lignin.

Studies on the use of natural fiber such as cotton, hemp, date palm, oil palm empty fruit bunch and wood flour as reinforcement in polyethylene, polyester, polypropylene and its recycled filled have been reported [3-5]. Besides cellulosic fiber, natural fibers are obtained from bacterial cellulose or microbial cellulose [6,7]. In laboratory scale preparation, bacterial cellulose is produced by an acetic acid-producing bacterium, *Acetobacter xylinum* through the fermentation process in the synthetic culture medium containing saccharides. Having its excellence properties such as high cellulose purity, high crystallinity, and high mechanical strength, bacterial cellulose exhibits promising alternative natural polymer for several applications.

Bacterial cellulose has been investigated for electronic paper displays [8], optically transparent composites [9], medical applications [10], and paper making [11]. Several synthetic polymeric materials have been reinforced with bacterial cellulose for composite preparation such as with thermoplastic starch [12, 13], phenol formaldehyde [14], polyethylene oxide [15] and polyester with rayon fiber [16].

Blending of polyethylene (PE) with plant natural fibers has been reported elsewhere [17,18]. However, no research on the use of bacterial cellulose in the blending with polyethylene has been reported. Polyethylene which is an important polymeric material due to its light-weight, low cost, good mechanical properties, and high thermal stability was used in this study. Therefore, this study aims to evaluate the effect of bacterial cellulose reinforced polyethylene on the physical and mechanical properties.

EXPERIMENTAL METHODS

Materials

Bacterial Cellulose (BC) gel was purchased from local small medium enterprise in Cianjur, West Java Province, Indonesia using coconut water as the main sources of saccharides enriched with nitrogen-containing compound. With the thickness about 1-2 cm, BCs were washed thoroughly by running tap water until its pH was neutral, then boiled with NaOH solution (2% w/v) for 1 hour to remove its impurities and to eliminate bacterial cells, and finally washed with water until its pH was 7. To reduce its excess water content, the wet BCs were squeezed to leave water content to 5-10 %. To obtain thin sheet of BC and to release its moisture excess, these BC was then pressed at a closed aluminum mold size of (270 x 270 x 4) mm with 0.3 mm thick spacer. The mold is then pressed at 50 kgf/cm² MPa and at 120 °C for 24 hours. Prior to

compounding, the obtained dry BC sheet was shredded and run through a ball mill to pass a 100 mesh screen, dried in an oven at 100 °C for 24 hours. Its moisture content was controlled within 2-3% and used as filler.

Since Maleic Acid Anhydride grafted Polyethylene (MAPE) has been demonstrated to be a successful coupling agent by creating polar or non-polar interactions between the matrix and fiber [19], thus MAPE was used in this experiment. MAPE with density of 0.92 g/mL at 25 °C and melting point of 107 °C was purchased from Sigma Aldrich and used as a modifier of BC/polyethylene composites.

Preparation of Polyethylene-Bacterial Cellulose Composite

Polyethylene (PE) and MAPE (2% w/w) were loaded into the mixer chamber at 180 °C processed at 60 rpm for 3 minutes. Dried particle BC was then added and mixed at 60 rpm for 30 minutes. The content of bacterial cellulose subjected to polyethylene was set from 0 to 50 (w/w)%. The composites were then molded into sheet by hot pressing. The prescribed amount of blended PE/MAPP (14-17) g was placed between a pair of glossing plate (11 x 11) cm with 1 mm thick spacer. The temperature of hot press was set at 180 °C and subjected to 7.5 MPa for 30 minutes followed by cold pressing at the same pressure for 10 minutes, the sheet was then cooled at room temperature.

Mechanical Properties Testing

Prior to mechanical properties test, samples were shaped to dumbbell according to ISO 527 type 5A then conditioned at 23 °C, RH 50% for a minimum 40 hours before testing. Tensile test was performed according to ISO 527 on a Universal Testing Machine (Orientec UCT-5T) using 5 specimens at room temperature (23 °C, 50 %RH) at tensile speed of 10 mm/minutes.

Water Absorption

Specimens of (50 x 25 x 3) mm of size were prepared from composites. Water adsorption test were conducted by submerging the specimens in water for 24 hours and measuring the increase in weight and thickness swelling as compared to the original oven dry weight of the specimens [20]. Five specimens of each type were tested in a adjusted room (23 °C and 50 %RH) and the results averaged. Water absorption was calculated as indicated by the Equation (1):

$$Water\ Absorption\ =\ \frac{W_1 - W_0}{W_0}\ x\,100\% \quad ... \quad (1)$$

Where :
W_1 = Weight of specimens after 1, 7, 21, and 30 days

of immersion

W_0 = Weight of specimens before immersion

Scanning Electron Microscope

Scanning Electron Microscope (SEM) machine JEOL Model JSM-6360LA was used to study the cross-sectional area of tensile fracture specimen. The specimen was cut and prepared under liquid nitrogen and mounted to an aluminium holder with double-sided carbon tape, then sputter coated with a thin gold layer and observed at an acceleration voltage of 20 kV.

X-Ray Diffraction

X-Ray Diffraction (XRD) spectra were detected using scintillation counter and a pulse height analyzer. CuKα radiation (wavelength of 0.15418 nm) was produced at 40 kV and 30 mA, produced by a Geiger Flex DXG2 (Rigaku Denki Co., Ltd.)

RESULTS AND DISCUSSION

Mechanical Properties

Mechanical properties are necessary for the end use of composite. Therefore, the effect of bacterial loading on the composite mechanical properties is determined. The variation of tensile strength, break strain and modulus of elasticity of polyethylene-bacterial cellulose composites as a function of bacterial cellulose fiber load is presented in Figures 1, 2 and 3 respectively.

It is clearly seen that tensile strength, break strain, and modulus of elasticity were significantly influenced by bacterial cellulose loading. As compared to polyethylene (bacterial cellulose content 0 %), the addition of bacterial cellulose into polyethylene matrix resulted in linearly decreasing of both tensile strength and break strain value, but increases modulus of elasticity. The higher the bacterial cellulose content, the higher the reduction of tensile strength and break strain value. When tested at 23 °C 50 % RH, tensile strength of PE was 21.2 MPa, break strain of 270 %, and modulus of elasticity of 763 MPa. With the BC loading up to 50 %, tensile strength dropped to be half of its previous value and break strain to be less than 0.5 %.

However, Young's modulus increased significantly to be more than 230 %. The decreasing in tensile and break strain property of composites (Figures 1 and 2) may caused by the poor fiber dispersion in matrix. The ultrafine ribbons of bacterial cellulose form a dense reticulated structured which is stabilized by the strong inter fiber hydrogen bonding makes bacterial cellulose fiber hardly dispersed each other easily, as bacterial cellulose fiber content increases. Therefore, bacterial cellulose was unable to withstand the high load leading to fractures loading. In addition, at the higher
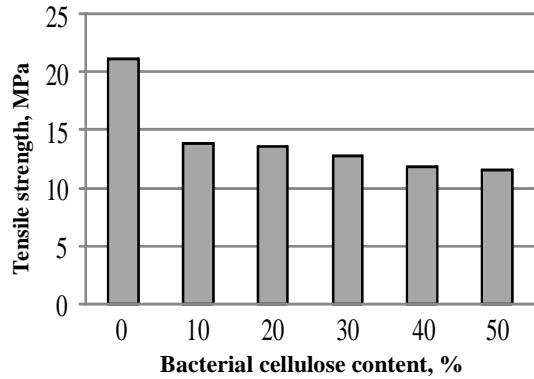

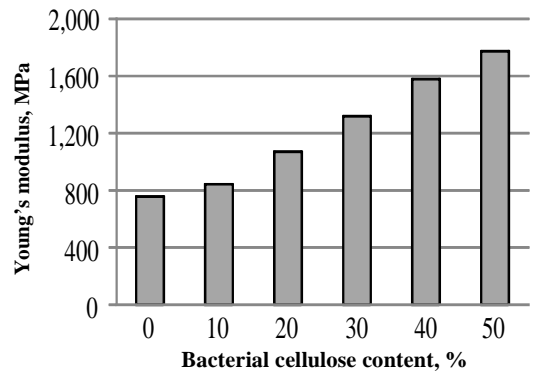**Figure 1.** Tensile strength of polyethylene composite with different concentration of bacterial cellulose.

Figure 2. Young's modulus of polyethylene composite with different concentration of bacterial cellulose.
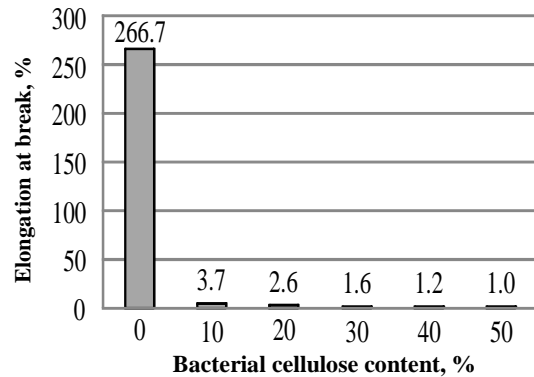

Figure 3. Elongation at break of polyethylene composite with different concentration of bacterial cellulose.

fiber loading, the process of fiber agglomeration becoming more difficult which in turn resulted in the void formation. This void affects the composite performance and lowered the tensile strength [21].

Other possibilities are due to the highly hydrophilic nature of bacterial cellulose and the hydrophobic polyethylene matrix. Hydrophilicity of bacterial cellulose is due to its polarity caused by the free hydroxyl groups derived from highly cellulose content (95 %) which may lead to poor bonding quality between fiber and matrix [22]. The less tensile strength of composites was also may due to the bacterial cellulose size caused in the less homogeneous

dispersion into the polyethylene matrix and less contact with polymer matrix, resulted the composite with lower tensile property [23].

Figure 2 shows that the addition of bacterial cellulose declined break strain of composites significantly, indicating that composites becoming more brittle by the bacterial cellulose loading. On the other hand, significant modulus of elasticity increased significantly as bacterial cellulose added (Figure 3). This caused by the fact that bacterial cellulose much more rigid than polyethylene matrix.

Water Absorption

Figure 4 shows the water absorption value of bacterial cellulose filled polyethylene. The values of water absorption of polyethylene which is hydrophobic in nature was negligible since it did not absorb water during 24 hours of immersion time. Therefore, it can be assumed that all the water absorption of the composites was caused by the bacterial cellulose and not by the hydrophobic polyethylene. Bacterial cellulose is very hydrophilic; the water molecule is easily absorbed into the composite. As bacterial cellulose was incorporated into the polyethylene, the composite was become more hydrophilic. Therefore, the composite was enabled to adsorb and retain more water than polyethylene. It indicates clearly that water absorption of composites increased with increasing amount of bacterial cellulose fiber. This is due to the hydrophilic nature of bacterial cellulose having cellulose containing free hydroxyl groups. These groups absorb water easily. The amount of hydroxyl groups is definitely caused by the high cellulose content in bacterial cellulose, approximately 95 %. These hydroxyl groups can take water molecules easily through hydrogen bonding in the fiber cell wall. Therefore, the higher the bacterial cellulose content in the composites, the higher the hydroxyl content, the higher water absorption [24].

Figure 4 shows that the addition of bacterial cellulose up to 50 % resulted in the highest value of water absorption. On the other hand, the decreasing amount of bacterial cellulose, the quantity of hydroxyl groups in composites decreases and leads to the
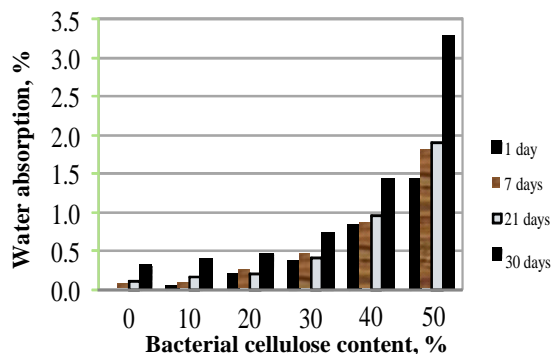
lowering water absorption. The natural water uptake ability of bacterial cellulose also makes it easier to reduce the physical bonding with matrices. In addition to the phenomenon of hydroxyl groups based water absorption ability, the increased absorption of composites at higher amount of bacterial cellulose can be attributed to the poor compatibility between the bacterial cellulose and polyethylene matrix. As the amount of bacterial cellulose increased, micro-level processing of the composites become difficult and might creates micro-void and cracks within the composites, caused the flow of water molecules along the bacterial cellulose - matrix interface, resulted in the diffusion from the interface into the matrix and fibers [25].

Scanning Electron Microscope

In order to show the fracture surface of BC/polyethylene composite before and after tensile test, morphology observation of cross-sectional using SEM was conducted to the composite containing 40 % bacterial cellulose as a representative of BC/polyethylene composites (Figure 5). It reveals that layered structure of bacterial cellulose fiber and voids in the composite are clearly visible (Figure 5(a)). Voids formation indicating the poor fiber-matrix bonding in composite . It has been known that the nature of matrix and the adhesion between fiber and matrix play an important role in tensile strength. The voids formation declined tensile strength as bacterial cellulose loading increases. For bacterial cellulose with high content of hydroxyl groups, the addition of 2 % of maleic anhydride polyethylene may
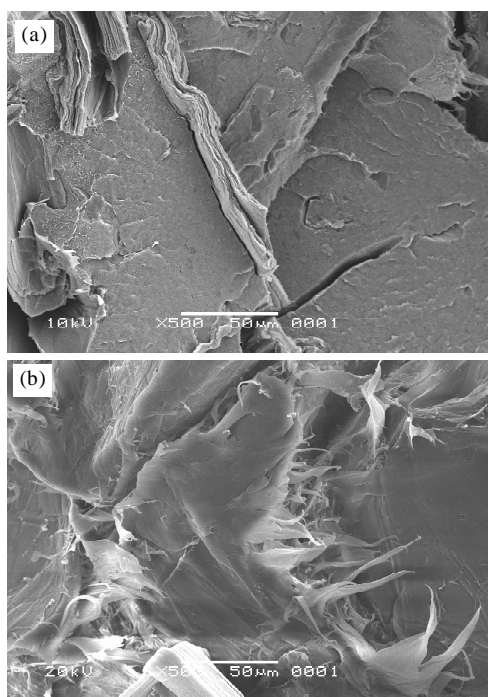

Figure 4. Water absorption of polyethylene composite with different concentration of bacterial cellulose.

Figure 5. Scanning electron micrographs of polyethylene composite with 40 % of bacterial cellulose content (a). before tensile testing and (b). after tensile testing.
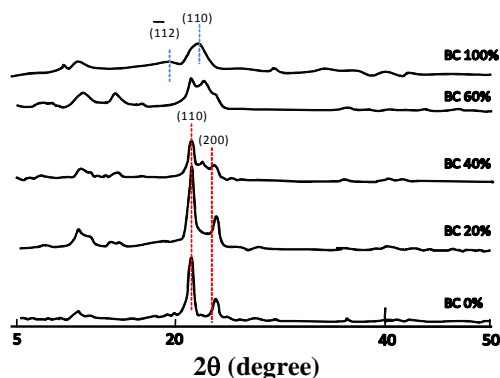
Figure 6. X-ray diffraction profiles of polyethylene composite reinforced with different concentration of bacterial cellulose.

inadequate to improve compatibility between bacterial cellulose and matrix. Therefore, the improvement process of the polyethylene-bacterial cellulose to create polar or non-plar interaction in composite is still needed in the further investigation. Figure 5(b) of the fracture surface of polyethylene-bacterial cellulose composite shows fiber ends and the pull-out length is large indicating poor fiber matrix bonding.

X-Ray Diffraction

The X-Ray diffraction profile of polyethylene reinforced with different content of bacterial cellulose is shown in Figure 6. It clearly shows the strong peak at $2\theta = 21.22°$ of (110) and $23.58°$ of (200) dominant appear in BC 0 and 20% as description of crystalline region in polyethylene (PE) structure. The intensity of peak greatly decreases after the addition of BC 40 %. This result was due to the change of BC structure from crystalline becoming more amorphous by the ball milled treatment. Consequently, the composite shows more amorphous property with the greater content of bacterial cellulose. In the X-Ray diffraction profile of BC 100 %, it showed typically cellulose with the strongest peak at 22.56° as lattice diffraction (110) and the weaker peak at 17.6 ° as lattice diffraction (112). However the intensity of peaks in bacterial cellulose decreased because of the ball mill treatment.

CONCLUSIONS

This study shows the use and properties of bacterial cellulose in polyethylene thermoplastic composites. Different composition of the composites was studied in respect to the mechanical and physical properties. Tensile strength and break strain declined significantly with the bacterial cellulose loading, but Young's modulus increased. Scanning electron microscope reveals the existence of voids resulting poor compatibility between bacterial cellulose and polyethylene. Works are in progress to enhance the interfacial interaction between the two materials.

Based on the above results, it can be concluded that the addition of bacterial cellulose up to 50 % the tensile strength dropped to be half, the break strain to be less than 0.5 %, but Young's modulus increased to be more than 230 %. In addition, composite of polyethylene with bacterial cellulose became more hydrophilic and amorphous

ACKNOWLEDGEMENT

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