Green Polyurethane Nanocomposites from Soy Polyol and Bacterial Nanocellulose

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Abstract

With increased environmental concerns, fluctuations in oil prices and dependency on oil, there has been an emergence in the use of biobased polyurethanes prepared with polyols derived from plant oils, such as soybean oil. In this study, novel polyurethane materials were synthesized using polyols obtained from soybean oils. The polyurethanes were produced by reacting the polyols with polymeric isocyanate with an isocyanate index of 100 at 150 °C for two hours for complete curing. The mechanical properties of this biobased polyurethane were improved by incorporating novel nano size cellulose materials produced from bacteria. The source of the bacterial cellulose nano-fibrils was a commercially available food product nata-de-coco. A fine dispersion of the nanocellulose fibrils in biobased polyurethane matrix was achieved using a high speed homogenizer at 30,000 rpm, which was observed using field emission transmission electron microscopy and scanning probe microscopy. The average diameter size of the cellulose fiberils were determined to be 22 ± 5 nm by scanning probe microscopy observations. The flexural strength and flexural strength was improved even at 0.125 wt% bacterial cellulose concentration and the optimum nanocomposite was obtained with 0.25 wt% concentration due to good interaction of isocyanates and the cellulose. Dynamic mechanical analyses were consistent with the flexural test results in terms of modulus. The transparent thick nanocomposite samples shows one additional advantage of the nanocomposite technology.

Introduction

Polyurethane materials attract much attention due to their versatile applications with a wide variety of polyols and isocyanates available to synthesize polyurethanes. The polyurethane was first discovered in the 1940s and still there is an extensive research on this type of polymer to improve the properties and to synthesize new materials with new functional groups. Polyurethanes are currently used as films, foams, and rigid materials¹⁻³. The main application areas are mainly construction materials and in the automotive industry in the form of foams and rigid applications⁴. This family of polymers is also widely used in coating, and elastomers⁵⁻⁷. Furthermore polyurethanes are used for some biomedical applications as well⁸.

With the increased environmental concerns and fluctuations in oil prices, interest in biobased materials is rising. Plants offer alternative chemicals to petroleum based chemicals. Plant based chemicals are renewable and they are biodegradable as a consequence of their biological properties. Extracted plant oils have been used in polymer synthesis ^{9,10}. Castor oil is commonly used for the synthesis of polyurethanes due to high hydroxyl content⁹.

In recent years, soybean oils are being converted to soy polyols by introducing hydroxyl groups to the structure to be reactive for polyurethane synthesis¹¹⁻¹³. Soy polyols with different functional groups and different hydroxyl content are also being produced. Soy polyols offer new polyurethane formulations, but for commercial applications soy polyols are not fully utilized in the polyurethane formulations due to reduction in mechanical properties compared with the

petrochemical polyols¹⁴. There have been studies to optimize the polyurethane synthesis using soy polyols with different hydroxyl numbers^{15, 16}, and different amounts of isocyanates¹⁷.

The properties of soy polyol based polyurethanes can be improved with certain reinforcing phases. There have been studies showing the improved properties of polyurethane with the use of glass fibers and natural fibers^{14,18,19}. Among the reinforcing materials, cellulose fibers, especially nano cellulose fibers, are attracting much attention²⁰.

Cellulose can be obtained from a vast range of sources, including plant, tunicates and certain strains of bacteria. Cellulose nanofibrils prepared from tunicates and cellulose produced via bacteria inherently have dimensions in the nano-scale. Cellulose nano-whiskers can be obtained from all the above sources, most commonly through acid hydrolysis of the cellulose fibers²¹. Detailed investigations on the whisker production are still under extensive research especially from different plant resources²². Microfibrillated cellulose is generally obtained by the defibrillation of the cellulose by the use of high pressure homogenizers^{7, 23}.

Bacterial cellulose is obtained via biotechnological routes²⁴⁻²⁶ and is commonly produced from the *Acetobacter xylinum* bacteria²⁷. It has the identical structure compared to plant based cellulose²⁴. It was reported that it has elastic modulus as high as 114 GPa²⁸. Bacterial cellulose can be regarded as a pure form of cellulose in comparison with those from plant sources, in order to obtain cellulose from plant resources, the cellulose should be separated from lignin, hemicellulose, pectin and waxes in the plant.

In this study, we hypothesize that bacterial cellulose nano-fibrils can reinforce the soy polyol based polyurethanes significantly and this can lead to novel polyurethane products with enhanced properties. Currently soy polyol is partially used in the polyurethane industry due to the lower mechanical properties. The pure form of cellulose at the nanoscale can improve the properties of soy polyol based polyurethane. We attempt to improve the mechanical properties of soy polyol based polyurethane by incorporating bacterial cellulose nano-fibrils .

Experimental

Materials. Soy polyol was obtained from Arkema Inc. with a hydroxyl number of 166 g KOH/mg, 0.42 acid value, and 1144 cps viscosity. Soy polyol was specially produced for this research and the polyol synthesis conditions are proprietary. The polyurethane synthesis was conducted with a commercial isocyanate (polymeric diphenyl methane diisocyanate (pMDI)) obtained as a gift from Huntsman Chemicals under the trade name Rubinate M. pMDI had a functionality of 2.7, and % NCO of 31.2. Dibutyl tin dilaurate (DABCO T12) catalyst was obtained from Air Products. The bacterial cellulose nano-fibrils were obtained from *nata-decoco*, a commercially available product, CHAOKOH, coconut gel in syrup (Thep. Padung Porn Coconut Co. Ltd. Bangkok, Thailand); the purification and extraction procedure is detailed below.

Extraction and purification of cellulose nano-fibrils. Bacterial cellulose was extracted from *nata-de-coco* in batches of five jars (of net weight 500 g each). For each batch, the coconut gel content was rinsed three times with 5 L of de-ionized water and blended for 1 min using a laboratory blender (Waring Blender LB20EG, Christison Particle Technologies, Gateshead, UK). The blended bacterial cellulose was then homogenized at 20,000 rpm in 5 L of water for 2 min using a homogenizer (Polytron PT 10-35 GT, Kinematica, CH) and centrifuged at 14,000 g to remove excess water and heated in a 0.1M NaOH solution at 80 °C for 20 min to remove any remaining microorganisms and soluble polysaccharides. The purified bacterial cellulose was then

successively centrifuged and homogenized to neutral pH using de-ionsed water. A dry cellulose nano-fibril product was obtained by freeze-drying as follows. The cellulose dispersion was adjusted to a concentration of 4 mg/ml in water, which was then poured into polyethylene beakers in volumes of 250 ml which were then frozen in liquid nitrogen, after 2 h the frozen product was transferred to freeze-dryer (Edwards Modulyo freeze dryer, West Sussex UK) and the resultant freeze-dried nano-cellulose fibrils retained for later modification and inclusion in composite production.

Synthesizing Polyurethane and Polyurethane Nanocomposites. The polyurethane reactions were carried out with an isocyanate index of 1.00 with a small amount of catalyst in the reaction system (0.2 wt %). The polyol, catalyst and pMDI were mixed for 2 minutes and the mixture was poured into molds and the polymer was cured for 2 hours at 150 °C using compression molding creating 3.5 mm thick polymer sheets.

The nanocomposites of polyurethane were prepared using the following steps. Firstly, bacterial cellulose was dispersed in the soy polyol manually and then further dispersed in the polyol with the use of a high speed homogenizer (Fisher Scientific PowerGen Model 125) at 30 000 rpm in an effort to improve homogeneous distribution of the nano-fibrils. The homogenized product was then placed in vacuum oven at room temperature for 3 hours to remove any air bubbles that formed during the homogenization process. After this de-gassing step, the catalyst (0.2 wt %) and pMDI with isocyanate index of 1.00 was added, mixed for 2 minutes and the nanocomposite poured into the square molds with dimensions of 71*50*3.5 mm and cured for 2 hours at 150 °C with compression molding with a pressure of 50 psi. The preparation steps are shown

schematically in Figure 1a. The various weight fractions investigated (up to a maximum of 0.375 wt% nano-cellulose fibrils) and their respective sample codes are summarized in Table 1.

Characterization

Fourier Transform Infrared Spectroscopy (FTIR)

FTIR measurements were conducted with Thermo Scientific Nicolet 6700, with the ATR attachment of the instrument. The number of scans was 32 for each sample with a resolution of 4.

Mechanical Testing

Flexural strength was measured with Instron (3382) testing machine according to ASTM D790. The flexural samples sizes were 10*60*3.5 mm and the speed of testing was 14.08 mm/min. Izod notched impact strength was measured with Testing Machine Inc. (TMI) according to ASTM D256. The sample sizes were same as flexural testing. The pendulum used for the test was 0.5*0.5 ft.lbs.

Dynamic Mechanical Analysis (DMA)

DMAQ800 from TA instruments was used to measure the storage modulus and tan delta of polyurethane and nanocomposites in three point bending mode. The samples were heated from - 50° C to 100° C and a rate of 3 °C/min with amplitude of 15 µm and a frequency of 1 Hz.

Transmission Electron Microscopy (TEM)

The images were taken at 200 kV on the FEI Tecnai G2 F20 TEM with a Gatan 4k digital camera using the DigitalMicrograph software. The samples were sectioned (100 nm) mounted on

300 mesh carbon grids and post stained with aqueous 2% Uranyl acetate (10 min) and Lead Citrate (4 min) then 10% Uranyl acetate in methanol for 12 min.

Scanning Probe Microscopy (SPM)

Agilent 5500 scanning probe microscope with Olympus IX71 inverted optical microscope was used to observe the nano size bacterial cellulose fibers. Silicon nitride tip with spring constant of 0.6 N/m was used for the observations.

Results and Discussion

Fourier Transform Infrared Spectroscopy. The synthesis conditions for the polyurethane were optimized using the FTIR analysis. The FTIR peak, at 2271 cm⁻¹ is very important for the polyurethane synthesis. This corresponds to free, unreacted isocyanate peak in the polyurethane structure. There was no free isocyanate peak observed at 2271 cm⁻¹ showing successful polyurethane synthesis. The specific peaks are shown in the Figure 2 for the polyurethane. These peaks are 2900 cm⁻¹, 1690 cm⁻¹, and 1376 cm⁻¹ corresponding to C-H stretching, C=O stretching, and C-N bonding, respectively³⁰.

In Figure 2, FTIR spectra of the polyurethane nanocellulose composites are also shown. The FTIR peaks show that nanocomposites were successfully synthesized without any isocyanate peak at 2271 cm⁻¹. The cellulose peaks could not be observed as the concentration of the cellulose in the polyurethane matrix did not exceed 0.375 wt%. It is reported that for polyurethane lignin blend casted as film, the polyurethane FTIR peaks were not altered much and lignin peaks were not observed even at 9 wt% lignin concentrations³¹. For a nanocomposite at 0.375 wt%, it is not possible to separate specific peaks of cellulose peaks. The amount of

cellulose is so low in concentration that it can not be easily detected. This observation shows the importance of the nanocomposites as the properties of the matrix polymer changes significantly with a very small amount of the nano filler.

Flexural Test Results Flexural testing was applied for the materials under investigation due to the application areas of the polyurethanes as they are mainly subjected to bending and flexural loads. The measurements and properties of the neat polyurethane and the nanocomposites are summarized in Table 2. It was observed that the nano cellulose, even at very low loadings (less than 0.5 wt%) percentages are very effective in reinforcing neat polyurethane. The fine dispersion of the nanocellulose fibrils with the polyurethane was observed with the transmission electron microscopy studies and atomic force microscopy studies. The increase in the strength values was 100% for 0.250 wt % nanocellulose and the increase in the modulus values was 50 % for 0.375 wt% for the nanocellulose addition. The strength and modulus values very leveling off at 0.250 wt% and 0.375 wt% due to saturation of cellulose dispersion at these concentrations. The electron microscopy studies also confirm that cellulose dispersions are similar and properties are quite similar. The cellulose dispersion reaches a maximum. During the experiments 0.5 wt% and higher concentrations could not be prepared due to agglomeration. This leveling was reflected in the flexural test data as well.

Cellulose and lignocellulosic materials are quite well known for their hydrophilic properties^{24, 32-³⁴. For example, Blaker et al.²⁴ measured the water contact angle in air of bacterial cellulose at ^{11°}. They modified cellulose to be hydrophobic with silane coupling agents to create good nanocomposite foams via Pickering emulsion templating²⁴. By tailoring the hydrophobicity of bacterial cellulose nano-fibrils by esterification with organic acids, polylactide matrix nano-} fibrillated bacterial cellulose composites with improved properties have been produced³⁵. Lu et al.³⁴ also modified the hydrophilicity of the microfibrillated cellulose to be compatible with the epoxy resins with silane and titanate coupling agents observing that titanate coupling agents improved the properties. The incompatibility of the cellulosic fibers with the thermoplastic resins has been one of the obstacles of the natural fiber composites. A recent study on nanocellulose composites with polypropylene has shown that the surface modification was needed for the cellulose whiskers to prepare successful nanocomposites³³. So in this study, polyurethane was used as the matrix polymer which can be regarded as a more hydrophilic material due the prevalence of hydroxyl groups obtained from the polyols during the synthesis. Polyurethanes are generally hydrophilic materials. In one study, the polyurethane water contact angle was reported by Seydibeyoglu et al.³ as 67°. The good compatibility of the cellulose with the polyurethane helps to improve the properties of the neat polyurethane matrix significantly. Furthermore, an improved interaction between the polyurethane and the cellulose arises due to the isocyanate groups in the polyurethane. The isocyanate has been used as surface modifier for the cellulosic based composite materials since 1989³⁶. The bacterial cellulose and the isocyanates can also react in this nanocomposite preparation system but the content of the bacterial cellulose (less than 0.5 wt%) is so low that this reaction would not contribute significantly.

Another important finding of the study was that the bacterial cellulose could not be incorporated to the polyurethane matrix more than 0.5 wt%. During the preparation of the samples, mechanical properties and the TEM images confirmed this observation. When the cellulose was dispersed with the homogenizer in the polyol shown in Figure 1, the bacterial cellulose could not be dispersed above 0.5 wt% as the cellulose was thickening the polyol and the cellulose fibrils agglomerated; nanocomposites at this higher loading were therefore not prepared. Different

forms of cellulose have been used as a viscosity modifier for many years and recently the viscosity change with cellulose addition was shown by Kahng et al.³⁷ The nano size effect becomes very effective by the dispersion of the nanosized cellulose fibers in the polyol.

Furthermore the results are compared with the theoretical model of "Rule of Mixtures" which is the fundamental theory for composites especially for long fiber composites. The rule of mixtures is stated in the following equation,

$$E_c = E_m * V_m + E_f * V_f \tag{1}$$

where E_c is the modulus value for the composite, E_m is the modulus value of the matrix polymer, V_m is the volume fraction of the matrix polymer, E_f is the modulus value for the fiber and V_f is the volume fraction of the fiber.

In the calculations for this study, the weight fractions are converted to volume fractions and volume fractions used are 0.1, 0.2, and 0.3 corresponding to 0.125, 0.250, and 0.375 wt% respectively. In Figure 3a and 3b, the experimental data and the calculated data are plotted. In Figure 3a, the experimental data and the theoretical model data are plotted with the modulus value of the bacterial cellulose taken as 114 GPa based on previous published data²⁸. In Figure 3b, this time the experimental data and the theoretical model is plotted but this time the modulus value of the bacterial cellulose is taken as 12.9 GPa. This value is calculated value from back calculations of the 0.2 volume % nanocomposites during this study.

Figure 3a shows that the experimental values are in a good trend but the modulus value for the bacterial cellulose taken from the literature did not fit into the experimental data. This can be attributed to the model adopted to long fibers whereas in this study, the bacterial cellulose as

shown by TEM images are short fibers as they are broken during the homogenization process for the fine dispersion. Thus the orientation of the short fibers is much different than that for long fibers. The second factor is that the bacterial cellulose would be modified during the polyurethane synthesis being exposed to chemical interactions of the polyol and the isocyanates thereby reducing the strength of the bacterial cellulose. The third factor may be the interface between the polyurethane and the bacterial cellulose. Though the strength values increase, better interface could have improved the adhesion yielding a higher modulus value for the bacterial cellulose.

The rule of mixtures can be rewritten with the correction factor, k which corresponds to the difference in the modulus values due to reasons explained previously. Rule of mixture is generally used for long fiber composites and the correction factor is generally used to adopt the rule of mixture to short fiber composites.

$$E_c = E_m * V_m + k * E_f * V_f \tag{2}$$

From these calculations k value can be calculated as 12.9/114 = 0.113.

So after these observations, the curves were plotted by taking the bacterial cellulose modulus value as 12.9 MPa. Figure 3b shows the experimental data and theoretical model, overlaid. The results are very promising showing almost the same trend. The rule of mixtures could be used for the bacterial cellulose nanocomposites with the corrected modulus values showing the linear trend. Even the modulus value of bacterial cellulose obtained is much lower than theoretical values, it is high compared to many polymeric materials and it shows that the nano cellulose has a big potential for many different applications.

The reinforcement of the soy polyol based polyurethanes was done previously with the use of the glass fibers and natural fibers^{14, 18, 19}. They observed the load transfer from the polyurethane matrix to the fibers which is the essential mechanism for the composite materials. By this study, with small amounts of nanocomposites similar reinforcements could be observed with less than 0.5 wt%. Latere Dwan'Isa et al.¹⁸ observed that soy polyol based polyurethane tensile strength was improved from 0.7 MPa to 1.1 MPa with 30 wt % glass fiber. So with the use of the nanocellulose at a loading of 0.250 wt% similar improvements could be achieved and this will help to improve the properties of soy polyol based polymers.

Impact Strength. To the best of our knowledge, this is the first report in literature of the measurement of the impact strength of nanocomposites based on nanocellulose. The results for this study are summarized in Table 2. For the natural fiber composites, the natural fibers generally decrease the impact strength of the polymer matrices^{38, 39}. The impact strength is related to the crack propagation through the polymer matrix and the energy absorbing capacity of the polymer during the sudden impact loadings¹⁸. For the polyurethane matrices, the impact strength is high having a value of 82.76 J/m. The impact strength of the neat polyurethane was improved with the glass fibers with a similar polymer matrix due to the high strength properties and energy absorbing mechanisms of the glass fibers¹⁸. In this study, it was observed that the presence of bacterial cellulose acted to decrease the impact strength of the composite, as has been found for other natural fiber reinforced systems^{38,39}. Whilst the polyurethane matrix is a tough, rubbery polymer with high energy absorption, it was not possible to improve the crack propagation with the use of nanocellulose due to the low impact properties of cellulosic fiber properties.

Dynamic Mechanical Analysis. The modulus values were measured using dynamic mechanical analysis at elevated temperatures to observe the changes of the storage modulus values with changes in temperature. The observations were consistent with the flexural test results. The storage modulus values increased as the nano cellulose content increased (Figure 4a). The increasing trend was similar to the modulus values obtained flexural test results and the values were also in the same range of 100-150 MPa. The polyurethane modulus was measured as 100.1 MPa at room temperature by DMA.

With the use of polyol with a hydroxyl number around 160 mg KOH/g, the polyurethane becomes flexible or semi-flexible but not rigid¹⁸. To achieve rigid polyurethane polyol with OH number of 250 or higher mg KOH/g is needed¹⁸. The flexibility of the polyurethane decreases as the nano cellulose is incorporated to the system. This is reflected in the modulus values as well. The improvement of the modulus values were observed throughout the entire temperature scan.

The representative tan delta curve for the polyurethane is shown in Figure 4b. T_g was measured from the onset temperature and peak of the tan delta curve as -1.15 °C and 37.73 °C respectively. The onset value for determination of T_g is more appropriate as the polymer chains start to soften and segmental mobility begins. Figure 4c shows the combined tan delta curves for PU and nanocomposites. As the bacterial cellulose is introduced into the polyurethane, the height of tan delta curves is reduced due to higher modulus values as tan delta is the ratio of loss modulus over storage modulus. The increase in the storage modulus results in decrease of the tan delta curves. There is a also shift in T_g values to higher values due to good interaction of the polyurethane and bacterial cellulose being both phases hydrophilic. The T_g value increases as the bonding between

the polyurethane and the cellulose makes the nanocomposite much more stiff resulting in higher glass transition temperatures.

Transmission Electron Microscopy and Visual Investigations of Samples. Obtaining images was problematic initially but with appropriate staining methodologies developed it was possible to observe the bacterial cellulose dispersions. The repeatability of the sample preparation techniques was confirmed by performing the experiment twice. Figure 5a, and 5b show the bacterial cellulose dispersions at different magnifications. Excellent distribution of bacterial cellulose nano-fibrils in the polyure than matrix is evidenced by the images. This is encouraging as it is generally very difficult to achieve good dispersion when producing nanocomposites. The fibrils are highlighted in the red circles (Figures 5a and 5b). The fine dispersion of the bacterial cellulose fibrils provides the increase in the flexural strength. The diameter of the bacterial cellulose fibrils are in the range of 20-30 nm which is consistent with the literature⁴¹⁻⁴³. Tokoh et al.⁴² and Astley et al.⁴³ estimated the thickness of bacterial cellulose around 30-50 nm. The otherwise fine dispersion of the nano cellulose fibrils in the polyurethane matrix was detrimentally affected at loadings above 0.375 wt%, as the fibrils tended to agglomerate, at these loadings good polyurethane nanocomposites could not be produced. Figure 5c shows the polyurethane nanocomposites for 0.250 wt% nano cellulose dispersion (the scale basr is 200 nm). The image can be compared with the Figure 5a and it can be easily concluded that the cellulose content is much higher than 0.125 wt% nanocomposites which is also reflected in the mechanical properties.

Figure 5d shows the polyurethane nanocomposites with 0.375 wt% nanocellulose. The fine dispersion was obtained. The images were similar to the images of PUBC250 which was very

important as the mechanical properties (flexural strength) were also leveling at these concentrations, corroborated by the TEM observations here.

Photographic images of the samples are presented in Figure 6, evidencing that all the samples including neat polyurethane and nanocomposites are transparent; one of the most important aspects of the nanocomposites^{44, 45}. At certain thicknesses and if the filler is well dispersed, transmission of light is not affected and not scattered so it passes through the material and that is why the nanocomposites with less than 100 nm thick fillers are transparent⁴⁵. The transparency was slightly reduced with bacterial cellulose compared to neat polyurethane but transparency of the nanocomposites was in the same range. The transparency of the products was observed with thick polymer samples of 35 mm, which is very novel. This observations is different from transparent nanocomposite materials based on films^{44,45}.

Scanning Probe Microscopy (SPM) Investigations. This study was conducted to observe the fiber structure at the nanoscale, resolved from 3D images obtained with the scanning probe microscopy. The images confirmed the TEM images obtained. The nano fibers at the range of the 20-30 nm were observed. Figure 7a shows the topography image of PUBC125. The nano fibers are shown with arrows. With the software, the average thickness of the cellulose nano fibers could be determined. The average thickness was resolved to 22.2 ± 5.3 nm (average based on 100 measurements), which shows the fine dispersion of the nano cellulose in the polyurethane.

Conclusions

Bacterial cellulose nano-fibrils were used to reinforce polyurethane, which itself was synthesized from 100% soy polyol. Nanocomposites with finely dispersed cellulose fibrils in the 16

polyurethane matrix were obtained. The properties of the polyurethane nanocomposites were investigated with Fourier transform infra-red spectra analysis, mechanical tests, thermal tests and microscopy techniques. It was observed that fine dispersion of the bacterial cellulose was achieved and as a result of both flexural strength and modulus improved over the unfilled samples by 100% and 50%, respectively. These significant reinforcements were achieved with less than 0.5 wt% bacterial cellulose in the polyurethane structure showing the importance of nanocomposites. The optical properties of thick sections of these nanocomposites were preserved.

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Figure Captions

Figure 1a: General Scheme for the Preparation of Polyurethane-Bacterial Cellulose Nanocomposites

Figure 1b: Model for the Fine Dispersion of Bacterial Cellulose in Biobased Polyurethane

Figure 2: FTIR Spectra of Polyurethane, PUBC125, PUBC250, PUBC375

Figure 3: Rule of Mixture Models Overlapped with Experimental Data

a) with literature modulus value, b) calculated modulus value

Figure 4: DMA Graphs of Polyurethane and Polyurethane-Bacterial Cellulose Nanocomposites, a) The combined storage modulus curve for PU and nanocomposites b) tan delta curve for the neat polyurethane, c) tan delta curves for PU and nanocomposites

Figure 5: Transmission Electron Microscopy Images, a) PUBC125 b) PUBC125, higher magnification, c) PUBC250, d) PUBC375

Figure 6: Digital photos of transparent nanocomposite samples

Figure 7: AFM Image PUBC125



Polyol-Bacterial Cellulose Before Mixing Hand Mixing



Polyol-Bacterial Cellulose After Hand Mixing ↓Homogenizer



Polyurethane Bacterial Cellulose Nanocomposite

Figure 1a

Compression Molding



Polyol-Bacterial Cellulose After Homogenizer



Figure 2







Figure 3b







Figure 4b



Figure 4c



Figure 5a



Figure 5c



Figure 5d



Figure 5b



PU





Figure 6





Table Captions

Table 1: Bacterial cellulose nano-fibril loading in polyurethane matrices and their corresponding sample codes

Table 2: Flexural strength, flexural modulus and impact strength of polyurethane and polyurethane-bacterial cellulose nanocomposites

Table 1

| Material | Bacterial Cellulose Content (wt %) | |
|----------|------------------------------------|--|
| PU | - | |
| PUBC125 | 0.125 | |
| PUBC250 | 0.250 | |
| PUBC375 | 0.375 | |

| Table 2 |
|---------|
|---------|

| Material | Flexural Strength (MPa) | Flexural Modulus (MPa) | Impact Strength (J/m) |
|----------|-------------------------|------------------------|-----------------------|
| PU | 3.03 ± 0.62 | 103.48 ± 31.28 | 82.76 ± 5.73 |
| PUBC125 | 3.81 ± 0.22 | 125.03 ± 80.05 | 52.08 ± 2.38 |
| PUBC250 | 6.00 ± 0.21 | 135.18 ± 11.49 | 48.29 ± 8.97 |
| PUBC375 | 5.28 ± 0.30 | 151.89 ± 20.94 | 42.13 ± 3.04 |