

MECHANICAL AND THERMAL PROPERTIES OF BACTERIAL CELLULOSE REINFORCED WITH BAMBOO MICROFIBRILLATED CELLULOSE AND PLASTICIZED WITH EPOXIDIZED WASTE COOKING OIL

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This paper aims to develop bacterial cellulose (BC) based biocomposites reinforced by bamboo microfibrillated cellulose (MFC) by the impregnation method, using epoxidized waste cooking oil (EWCO) as plasticizer, and to investigate the effect of EWCO on the properties of the biocomposites. The obtained materials were characterized by tensile testing, thermogravimetric analysis (TGA), X-ray diffraction (XRD), differential scanning calorimetry (DSC), scanning electron microscopy (SEM), and Fourier-transform infrared spectroscopy (FTIR) analyses. The optimum composition of the biocomposites was achieved with 1% w/w bamboo MFC as reinforcement agent and 0.5% v/v EWCO as plasticizer. Triple replications of the tensile test revealed slight differences between the unplasticized BC-based biocomposites and the plasticized BC-based biocomposites. The tests revealed that MFC impregnation in the BC-based biocomposite can enhance tensile strength. Thus, while the tensile strength of the BC was 9.6 MPa, it reached 40 MPa for the unplasticized BC-based biocomposites reinforced with bamboo MFC, and 37.8 MPa for the plasticized BC-based biocomposites. Reinforcing with bamboo MFC and plasticizing with EWCO can increase the degradation temperature of the BC-based biocomposite, while plasticizing can affect the crystalline structure of the material. Both BC-based biocomposites reached higher glass transition temperature than that of the initial BC.

Keywords: bacterial cellulose, microfibrillated cellulose, bamboo, epoxidized waste cooking oil, plasticizer

INTRODUCTION

The use of bacterial cellulose (BC) in the development of biocomposites is expected to yield reliable and environmentally benign materials. Previous research has demonstrated that BC possesses special and unique properties that differentiate it from plant-derived cellulose.¹ Recently, this type of cellulose has gained much attention due to its superior properties, such as high purity and biocompatibility.² Moreover, it is considered as a valuable component in composites, which by definition, represent the combination of two or more elements that form together a new material, with enhanced properties, compared to those of the individual components.³

BC can be produced by *Mycoderma aceti*, also known as the mother of vinegar.⁴ Several studies investigated the potential of various bacteria in BC production,^{5,6} including *Bacillus amyloliquefaciens*⁷ and *Acetobacter xylinum*.^{8,9} Thus, *Acetobacter xylinum* (*Gluconacetobacter*

xylinum) is considered the most efficient producer, accepting a wide range of carbon/nitrogen sources.^{10,11} BC that grows in coconut water is popularly used as dietary fiber (*nata de coco*) in Thailand, Malaysia, and Indonesia, where it is sold in the form of gelatinous cubes.

BC-based biocomposites can be developed by the addition of organic or inorganic molecules, including polymers, metal oxides, solid materials, nanomaterials, used either as matrix or as reinforcement.¹³ Such composites have been found suitable for application in several fields, such as electronics, biomedical engineering, food, biodegradable packaging *etc.*¹²⁻¹⁷ For example, previous work has reported on the successful use of a composite film from thermoplastic starch and bacterial nanocellulose as an alternative wrapping material to prolong the shelf life of minimally processed mangoes.¹⁷

Considering the advantages of BC, including its properties and relatively simple isolation and purification procedures, which do not require intensive energy or chemical processes,¹⁸ this study aimed to prepare biocomposites based on BC as a matrix, reinforced with bamboo microfibrillated cellulose (MFC) and plasticized using epoxidized waste cooking oil (EWCO). The objective was to obtain an environmentally friendly material by using bamboo as raw material based on sustainable locally available resources, as well as waste cooking oil (WCO) as a bioplasticizer, which is known for its potential to cause environmental pollution problems because of improper disposal.

Significant previous research has been made on the use of MFC as reinforcement in various composites, for example, by dispersing MFC in a polylactic acid (PLA) matrix, using acetylated bamboo MFC to toughen the PLA matrix, in polyvinyl alcohol/starch reinforced by bamboo MFC, *etc.*¹⁹⁻²² The findings have shown that bamboo MFC dispersed with an ultrasonic homogenizer to reinforce sago starch-based bioplastics increased the tensile strength of the materials to 21.9 MPa, at 5 wt%-w loading.²³ In another study, a composite material developed from BC grown on yam bean, reinforced by 0.375 wt%-w bamboo MFC, yielded a tensile strength of 22.41 ± 7.34 MPa.²⁴

Other research works have reported that plasticizers can reduce polymer-polymer interactions, imparting flexibility, moisture resistance, and ease of processability.²⁵ Various kinds of vegetable oils have been used as plasticizers, including palm oil olein,²⁶ rubber seed oil, Madhuca and Neem oils,²⁷ cotton seed oil,²⁸ palm kernel oil,²⁹ *Jatropha curcas* oil,³⁰ soybean oil,³¹ and mahua oil.³² Given the huge amount of waste cooking oil generated per year, of approximately 29 million tons,³³ it has been considered as a potential source of bioplasticizer to enhance the performance of biocomposites. It has been established that adding EWCO as a plasticizer during the preparation of sago starch-based biocomposites reinforced with bamboo MFC significantly increased the tensile strength and the thermostability of the materials.³⁴ Also, epoxidized soybean oil has been treated by tannic acid and used in the formulation of biocomposites reinforced with microfibrillated cellulose.³⁶ Moreover, epoxidized soybean oil has already been considered for the synthesis of lubricants through a process that can be integrated within

existing biodiesel plants.³⁵ The performance of the epoxydation method of WCO was investigated using with different inorganic acids to achieve optimum conversion.^{37,38}

Considering the positive results of previous studies that reported on the use of modified BC in combination with different plasticizers, such as glycerol and sorbitol, with various fillers, and achieved composite materials with increased tensile strength,³⁹ or various functionalized materials,⁴⁰⁻⁴² this work aimed to prepare BC-based biocomposites reinforced with bamboo MFC and plasticized with EWCO. The biocomposites were manufactured by the impregnation of the BC with EWCO and bamboo MFC. This study aimed to find out the optimum formulation for the biocomposites, varying the loading of the bamboo MFC, as well as examining the effect of plasticization using EWCO on the properties of the achieved materials.

EXPERIMENTAL

Materials

BC sheets were prepared on a laboratory scale, using sterilized coconut (*Cocos nucifera*) water as growth medium for *Acetobacter xylinum*, to which glucose and additional nutrients, such as MgSO₄ and KH₂PO₄, were added. Bamboo MFC and EWCO were prepared at the Advanced Material Laboratory of the Department of Chemical Engineering, Diponegoro University. The bamboo used belonged to *Dendrocalamus asper* species, obtained locally from a field in Sragen, Central Java, Indonesia. EWCO was prepared from waste frying oil filtered for removing solid waste. Hydrogen peroxide (50%), alcohol 96%, glacial acetic acid (CH₃COOH >99.9%), sodium hydroxide (NaOH), potassium iodide (KI), cyclohexane, nitric acid (HNO₃ >65%), Wijs solution, potassium hydroxide (KOH pellets), toluene, hydrochloric acid (HCl 37%), sulfuric acid (H₂SO₄ 96%), diethyl ether (99.6%), and sodium thiosulfate (Na₂S₂O₃ >>99.5%) were supplied from Sigma Aldrich, and used in this study without further purification.

Methods of preparation

Each culture medium consisted of 100 mL of coconut water, 2 g KH₂PO₄, 2 g MgSO₄, 1 g urea and 15 wt%-w glucose, into which 20 vol%-v *Acetobacter xylinum* was introduced and maintained at room temperature. The generated BC pellicles were harvested after 7 days and washed under running water. The samples were treated by immersing into 1% NaOH for 2 hours. The NaOH-treated BC pellicles were rinsed under running water to achieve neutral samples, which were denoted as BC.

The preparation of bamboo MFC samples was described in detail in a previously reported study.²³ The ground and screened bamboo was subjected to chemical treatments with ethanol and n-hexane, KOH and H₂O₂, to perform extraction, delignification and bleaching, respectively. The cellulose was washed with water, with vacuum assisted filtration, until neutral pH was achieved in the filtrate and then dried at a temperature of 105 °C until constant weight. Bamboo MFC was obtained by dispersing 1, 3, 5, 8 and 10 wt% -w in aquadest and subjecting the suspensions to an F250N ultrasonic homogenizer for 2 h.

The preparation of EWCO was also based on a previous study, and performed under optimum conditions.³⁴ The epoxidization reaction was performed by the addition of glacial acetic acid and hydrogen peroxide into waste cooking oil for 5 hours. In this reaction, sulfuric acid was used as a homogenous catalyst to obtain 3.1% oxirane conversion.

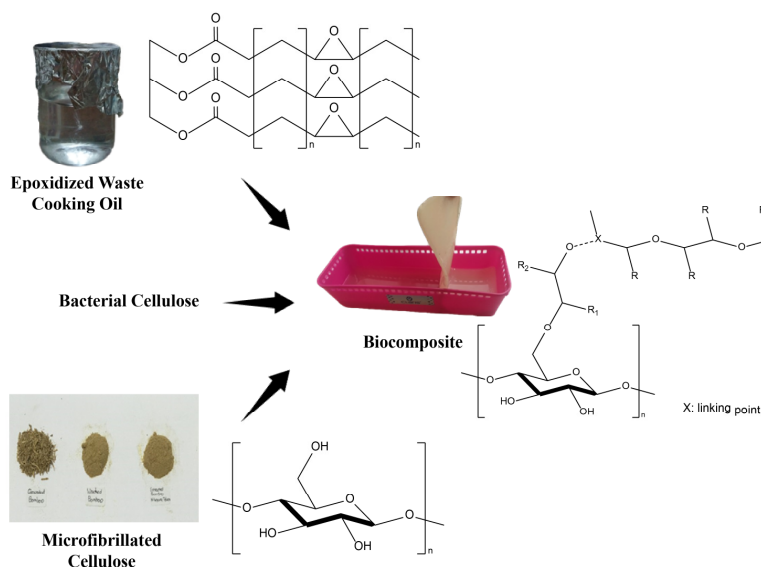
Different concentrations of MFC (1, 3, 5, 8 and 10 wt%-w) were used to prepare the BC-based composites in order to find the optimum formulation. MFC suspensions with different concentrations and 0.5 vol%-v EWCO were added to NaOH-treated BC sheets and left overnight. Some initial BC samples, without

the addition of MFC and EWCO, were used as controls. The thus-prepared materials were pressed using a laboratory hot press at a temperature of 7 °C for 60 minutes to obtain thin films.

Characterization

The tensile strength of the samples was determined using a CT3 Texture Analyzer, with a loading weight of 4500 g (AMETEK Brookfield, USA). Three repetitions of the tests were carried out on rectangular strips (5 mm x 20 mm) of both BC and BC-based biocomposite samples.

The BC-based biocomposites reinforced with bamboo MFC and plasticized with EWCO were subjected to DSC analysis, using a Shimadzu DSC-60 Plus (Japan) with inert nitrogen gas (N₂). Samples of each type, weighing 7.5 mg, were used for the DSC analysis. The samples were heated from 30 °C to 300 °C, at a rate of 10 °C/min, hold temperature of 300 °C, and N₂ flow rate of 30 mL/min, using an Al seal pan. The TG curves were recorded using a Shimadzu DTG-60 (Japan) under N₂ flow, at a flow rate of 30 mL/min, from 30 °C to 300 °C temperature, with a heating rate of 10 °C/min. FTIR analysis was performed on a Shimadzu IRPrestige-21 Spectrometer (Japan), at 4 cm⁻¹ resolution and with 40 scans.



Scheme 1: Graphical representation of the preparation of BC-based biocomposites

Also, the lateral order index (LOI) and hydrogen bond intensity (HBI) of the prepared materials were determined. HBI and LOI were calculated using the ratio of the absorbance bands at 1430 cm⁻¹ to 898 cm⁻¹, and 3400 cm⁻¹ to 1320 cm⁻¹, respectively.⁴³ The morphology of the BC-based biocomposites was observed using an Inspect S50 (FEI) SEM. The crystalline structure of the samples was analyzed using a PANalytical X'Pert Pro XRD (Utrecht, Netherland),

with Cu as anode material, in the 2θ range from 10° to 70°.

RESULTS AND DISCUSSION

The tensile strength of the BC-based biocomposites was found to increase with a loading of 1 wt% MFC-w (Fig. 1). Thus, as shown in Figure 1, the tensile strength of initial

BC was 9.62 ± 0.56 MPa, which was significantly increased after the addition of 1 wt%-w MFC. However, a rising MFC content affected the tensile strength of the materials. This can be explained by the good dispersibility of MFC when added in low loadings to BC.⁴⁴ On the other hand, as the content of the MFC filler rose, the tensile strength declined, probably because of poor interfacial adhesion between the filler and the matrix, which weakened the filler-matrix interaction.⁴⁸

Also, it can be observed that the tensile strength decreased for the plasticized BC-based biocomposites. Other studies on epoxidized vegetable oils also stated that the bioplasticizer can decrease tensile strength, as in the case of poly(lactic) acid plasticized with epoxidized palm oil (EPO) and epoxidized soybean oil (ESO).^{45,46} On the other hand, in another study, the authors reported on achieving microcrystalline cellulose reinforced polylactide acid composites, with the

addition of epoxidized soybean oil, with high toughness and improved tensile strength.⁴⁷

Considering the results for the tensile strength obtained in this study, the reinforcement with 1 wt%-w MFC was considered as the optimum in both unplasticized and plasticized BC-based biocomposites.

The data in Table 1 allow comparing the tensile strength results obtained in the present work with those of other studies reported previously. Thus, earlier research reported on BC-based biocomposites developed with poly(vinyl)alcohol (PVA) as matrix and the material reached a tensile strength of 37.9 MPa, significantly higher than that of the PVA alone.⁵¹ This value is very close to that achieved in our study for the plasticized BC-based biocomposite containing 1 wt%-w MFC. Lower tensile strength values have been obtained for other biocomposite formulations, such as the BC-Polyester⁴⁹ and BC-HEC composites.⁵⁰

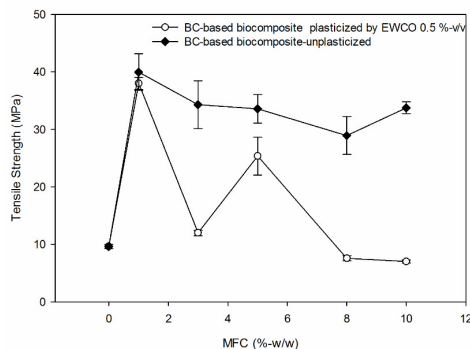


Figure 1: Tensile strength analysis of the BC and the BC-based biocomposites

Table 1
Tensile properties of BC and BC-based composites

Sample	Tensile strength (MPa)
BC	9.62 ± 0.56
BC-MFC (1 wt%-w)	39.97 ± 5.52
BC-MFC (3 wt%-w)	34.32 ± 7.20
BC-MFC (5 wt%-w)	33.59 ± 4.33
BC-MFC (8 wt%-w)	28.93 ± 5.69
BC-MFC (10 wt%-w)	33.76 ± 1.75
BC-MFC (1 wt%-w) -EWCO (0.5 vol%-w)	37.99 ± 1.77
BC-MFC (3 wt%-w) -EWCO (0.5 vol%-w)	11.98 ± 0.83
BC-MFC (5 wt%-w) -EWCO (0.5 vol%-w)	25.36 ± 5.71
BC-MFC (8 wt%-w) -EWCO (0.5 vol%-w)	7.55 ± 0.77
BC-MFC (10 wt%-w) -EWCO (0.5 vol%-w)	7.00 ± 0.47
BC-based materials from previous studies	
BC-Polyester ⁴⁹	20.5-26.7
BC-HEC ⁵⁰	12.7-15.4
BC-HEC-Polycyanate ⁵⁰	27.4-34.3
BC-PVA ⁵¹	37.9

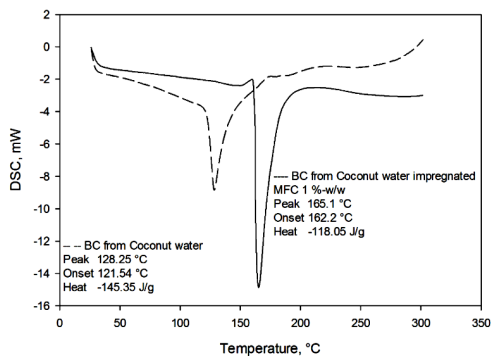


Figure 2: DSC thermograms of BC and BC-based biocomposite reinforced by 1wt%-w MFC

DSC and TGA analyses examined the thermal stability of the unplasticized and plasticized samples. The DSC analysis was used to obtain the glass transition temperature (T_g) of BC and both BC-based biocomposites. Figure 2 shows the DSC thermograph in the temperature range of 30 °C and 300 °C, with one peak assigned to the glass transition temperature. Upon reinforcing and plasticizing the BC, the glass transition temperature of the BC-based biocomposites reached higher values – from approximately 116 °C for BC, to 161 °C and 157 °C, for the unplasticized and plasticized samples, respectively, as shown in Figures 2 and 3. The incorporation of MFC into the BC matrix affected the molecular motion within the composites, causing a transition from the glassy state to the rubbery one. These results are important for determining the suitability of the prepared materials for specific applications, as well as their storage temperature, to avoid their degradation.^{52,53}

The thermal degradation behaviour of the composites was examined by TGA. The TGA results reveal the stages of thermal degradation. The samples underwent a small weight loss with a rising temperature from 30 °C to 100 °C. This was due to water evaporation, causing a weight loss of approximately 3% and 10% for the unplasticized and plasticized BC-based biocomposites reinforced with 1 wt%-w MFC, respectively. Meanwhile, a significant decomposition occurred at in the temperature range from 142 °C to 176 °C, with more than 90% weight loss being recorded for the materials. The DTG curves presented in Figure 4 indicate that the maximum rate of decomposition occurred at 150 °C for BC. Meanwhile, both BC-based biocomposites (unplasticized and plasticized)

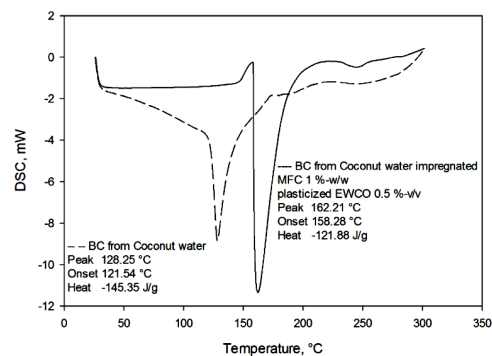


Figure 3: DSC thermograms of BC and BC composite reinforced with 1wt%-w MFC and plasticized with EWCO

recorded higher maximum decomposition temperature due to the incorporation of MFC. The results demonstrate that more energy was necessary for the composites to decompose. Also, the plasticized biocomposite reinforced with 1 wt%-w MFC underwent lower weight losses during its decomposition up to 300 °C.

Figure 5 shows the FTIR spectra in the 4000-400 cm^{-1} region of BC and BC-based biocomposites, where the typically BC bands can be observed.⁵⁴ Thus, the band at 3475 cm^{-1} is characteristic of O-H stretching of intra- and intermolecular H-bonds for cellulose I, that at 2900 cm^{-1} is assigned to C-H stretching, and that at 1620 cm^{-1} – to the bending vibration of naturally adsorbed water. The spectra of the BC-based biocomposites reinforced with MFC show the same bands, but with different intensity. As has been suggested earlier, the oxirane ring can be detected in the wavenumber ranges of 750-880 cm^{-1} and 815-950 cm^{-1} , while epoxides absorb near 1250 cm^{-1} .^{26,34}

The changes in the crystallinity order of cellulose can be noted from the HBI and LOI results listed in Table 2. The HBI of BC slightly decreased with the reinforcement with 1 wt%-w MFC, but increased back when the BC-based biocomposite was plasticized, exceeding that of the initial BC. As for the LOI of the BC-based biocomposites, an increase by 12% is noted for the MFC reinforced composite plasticized with EWCO. It appears that the addition of MFC further contributes the organized and ordered cellulose structure, leading to a higher hydrogen bonding. This conclusion was also supported by the thermal stability results, which demonstrated an increase in the decomposition temperature.⁴³

The morphological characteristics of BC and BC-based biocomposites were observed by SEM.

The micrographs shown in Figure 6 reveal the fine surface of BC with a particle length of 5-10 μm (A), the MFC (fibre length of 10 μm) covered

surface of BC (B), and the smoother surface of the biocomposite reinforced with MFC due to plasticization with EWCO (C).

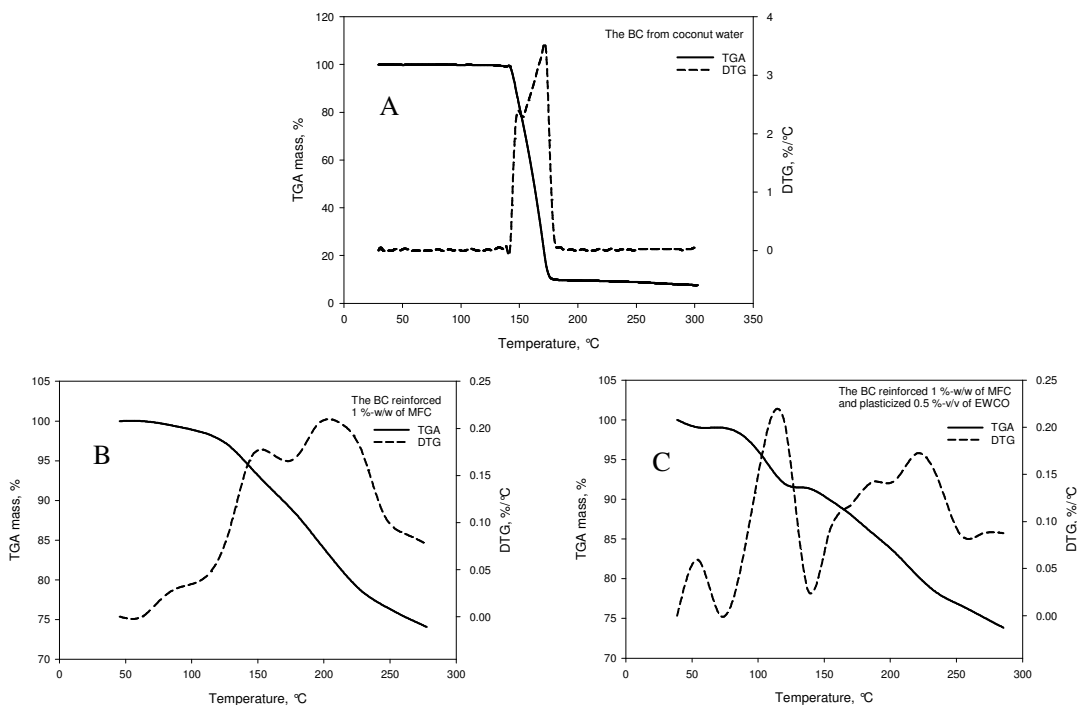


Figure 4: TGA and DTG thermograms of: (A) BC, (B) unplasticized BC reinforced with 1 wt%-w MFC, and (C) BC reinforced with 1 wt%-w MFC, plasticized with 0.5 vol%-v EWCO

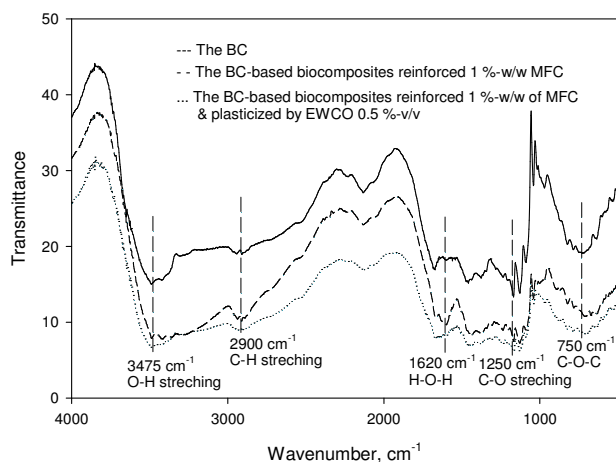


Figure 5: FTIR spectra of BC, unplasticized BC reinforced with 1 wt% MFC, and (C) BC reinforced with 1 wt%-w MFC and plasticized with 0.5 vol%-v EWCO

Table 2
HBI and LOI values determined for BC and BC-based composites

Material	HBI	LOI
BC	0.86	0.60
Unplasticized BC-based biocomposite	0.81	0.60
Plasticized BC-based biocomposite	0.88	0.67

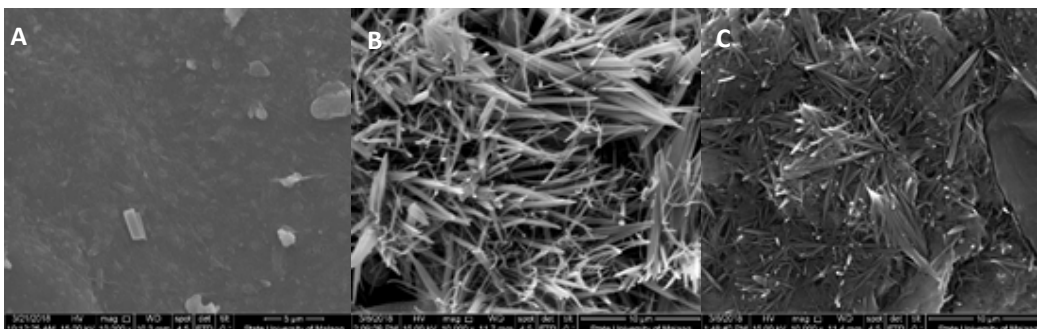


Figure 6: SEM images of BC (A), unplasticized BC-based biocomposite reinforced with 1 wt%-w MFC (B), and BC-based biocomposite reinforced with 1 wt% MFC and plasticized with 0.5 vol%-v EWCO at 10,000x magnification

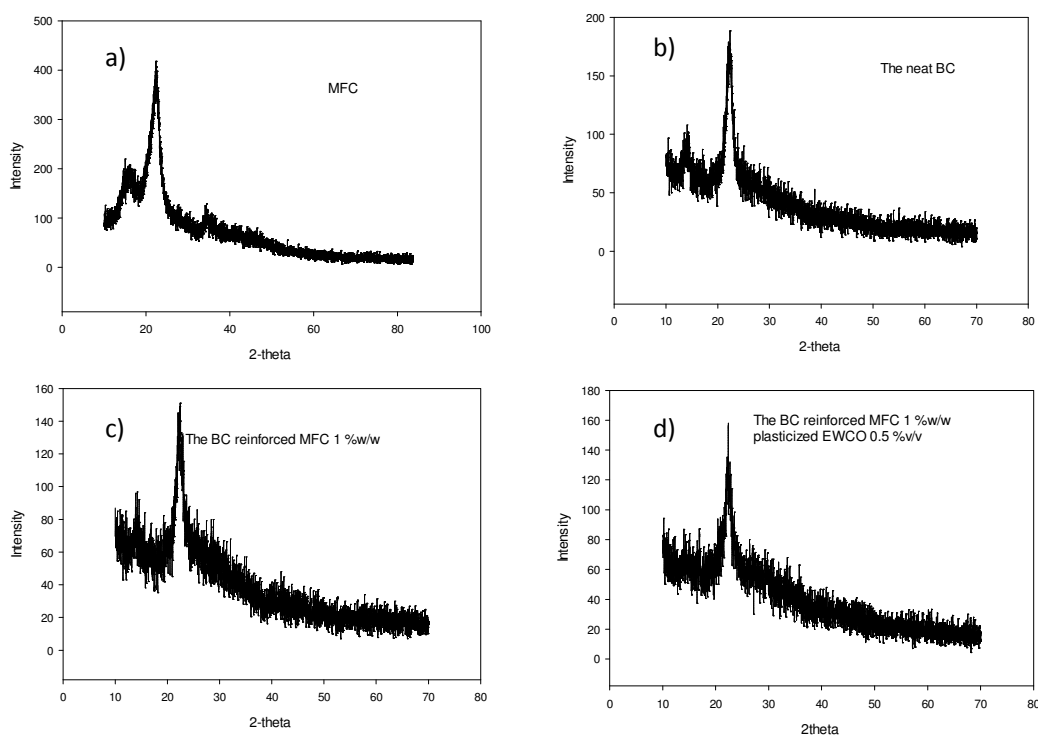


Figure 7: XRD patterns of: a) MFC; b) BC; c) unplasticized BC-based biocomposite reinforced with 1 wt%-w MFC; d) BC-based biocomposite reinforced with 1 wt% MFC and plasticized with 0.5 vol%-v EWCO

Figure 7 presents the X-ray diffraction patterns of the initial MFC and BC, as well as those of the BC-based biocomposites reinforced with 1 wt% MFC, both unplasticized and plasticized. XRD results give a better understanding of the crystalline structure of BC. As stated previously, high intensity peaks in the diffraction plane around 14.5° and 22.6° exhibit the cellulose I structure.⁵⁵ In the present study, the XRD patterns

of MFC and BC, as well as that the BC-based biocomposite reinforced with MFC, reveal two significant diffraction peaks at 14° - 15° and 22 - 23° , which confirms the presence of cellulose I structure. However, the peak at 14.5° disappeared in the pattern of the plasticized BC-based biocomposite, showing that the plasticization reaction affected the crystalline structure of the samples.

CONCLUSION

In the present work, bacterial cellulose (BC) based biocomposites, reinforced by bamboo microfibrillated cellulose (MFC) and plasticized using epoxidized waste cooking oil (EWCO), were prepared and their properties were investigated. FTIR, DSC, TGA, XRD and SEM techniques were used to characterize the obtained materials. The results showed that the composites reinforced with 1 wt%-w MFC showed higher tensile strength and thermal stability, which were maintained after plasticizing with EWCO. This study thus demonstrates the possibility to develop an environmentally friendly material, using raw materials from sustainable sources, with high tensile strength and thermal stability for various applications.

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