

## High barrier green food packaging materials from composites hybridization

<sup>1,2,\*</sup>Zakaria N.E., <sup>2</sup>Ahmad, I., <sup>1</sup>Busu, W.N.W., <sup>1</sup>Ismail, N., <sup>3</sup>Mohamed, W.Z.W. and <sup>2</sup>Baharum, A.

<sup>1</sup>Food Design, Processing and Packaging Program, Food Science Technology Research Centre, Malaysia Agricultural of Research and Development Institute, P.O Box 12301 General Post Office, 50774 Kuala Lumpur, Malaysia

<sup>2</sup>Centre of Advanced Material and Renewable Resources, Faculty of Science and Technology, National University of Malaysia, 43600 UKM Bangi, Selangor, Malaysia

<sup>3</sup>Faculty of Engineering and Life Sciences, Universiti Selangor 45600 Bestari Jaya, Selangor, Malaysia

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### Abstract

This research was done to study the effects of chemical modification on the mechanical, thermal and hydrophobic properties of treated *Sansevieria trifasciata* fibre (STF) in hybridized polymer composites. Processing of the composites was done by using an internal mixer machine. The processing parameters used were 135°C for temperature and a mixing rotor speed of 55 rpm for 15 mins. Filler loading was varied from 10% to 40% of STF and the fibre size used was 125 µm. The composite blends obtained then were pressed with a hot press machine to get samples for testing. Samples were evaluated via tensile tests, water absorption tests, water contact angle tests, antioxidant activity and scanning electron microscopy (SEM). Results showed that chemical modification by using silane-coupling agent has improved tensile strength, strain values and decreased water absorption of the treated composites. Antioxidant activity in treated fibre was also higher than untreated fibre, due to the removal of the impurities during the treatment. SEM micrographs have proven that the treated fibre has produced a rougher surface for more inter-facial interactions between fibre and matrix. Treated composites have given higher contact angle values compared to untreated composites and polymer matrix, contributed by the increment of the hydrophobic surface of the materials. From the results obtained, the treated composites have the potentials to be applied as packaging films to prolong the shelf life of food products due to their high barrier properties to water and moisture, even higher than typical packaging plastic (i.e HDPE). Materials obtained are also suitable for natural antioxidant packaging film and biodegradable packaging applications.

## 1. Introduction

Plastic waste disposal is a global issue. According to Dahlbo *et al.* (2018), within 50 years (1964-2014), plastic production all over the world had increased from 15 million tons to 311 million tons. In Europe, packaging industries are the main user of plastics (39.9%) in 2015, followed by the construction sectors. Developing new technologies and methods to treat solid wastes is a worldwide problem, especially for non-naturally-reversible polymers. Producing environmentally friendly (green) materials is a good choice to encounter problems, especially in packaging industries. Many types of filler have been used in the plastic industry, either to cut production costs or to tailor the mechanical properties of the materials. Hybridization of composites by combining

two or more fillers with a polymer matrix is a good effort in order to produce materials with better or additional properties.

Hybridization is done to improve either mechanical, thermal, damping, permeability or other additional properties for better performance of materials. These few decades, incorporating natural filler into polymer matrix has gained a lot of attention from research groups all over the world. There are many types of filler either natural or synthetic that have been used in the polymer industry either to cut production costs or to tailor the mechanical properties of the materials. Nowadays, researchers are keener to explore natural fibre filler due to the attractive features offered including their

\*Corresponding author.

Email: [ezdiani@mardi.gov.my](mailto:ezdiani@mardi.gov.my)

lightweight, low density, recyclable, moderate strength, high specific modulus, renewable, biodegradable, lack of health hazards, eco-friendly material, amenability to chemical modification and can be obtained at low cost (Ku *et al.*, 2011; Sreenivasan *et al.*, 2011; Faruk *et al.*, 2012). There are many types of natural fibre from plant resources. Most of them are agro-waste. Natural fibres like flax, hemp, jute, sisal, kenaf, coir, kapok, banana, henequen, rice husk, pineapple leaf, oil palm, wood pulp and many more are commonly used as reinforcement in polymer matrixes (Sreenivasan *et al.*, 2011; Pickering *et al.*, 2016). Therefore, new fibre resources should be explored. There are many fibre-yielding plants in the world that can offer a lot of potential to composite industries.

*Sansevieria trifasciata* (ST) are usually used as decoration plants at the park or at home. *Sansevieria* plants can also grow in wild environments (Sreenivasan *et al.*, 2011). Common names for ST are snake's tongue, devil's tongue, mother in laws tongue, bowstring hemp or snake's plant. ST is a type of herb with 2-6 leaves arising from the underground rhizome. The leaves have zebra-like markings which are erect, sword-shaped and leathery with cross-banded dark and light yellowish-green shades of 0.3-1 m long. Figure 1 shows the ST tree that has produced a 1 m long leaf. ST can grow anywhere anything, in full sun or light shade but thrives well in moist, fertile soil with high organic content with minimum care (Kanimozhi, 2011). Current research showed that utilizing *Sansevieria trifasciata* fibre (STF) as reinforcement filler in composites is still at the preliminary stage. STF has a lot of potential to be explored. It is observed that *Sansevieria* fibres have good mechanical and thermal insulation properties (Ramanaiah *et al.*, 2013).



Figure 1. *Sansevieria trifasciata* tree with 1 m long leaf.

Adding natural rubber (NR) into the composite blend also will modify its characteristics. It was reported that better tensile properties would be obtained when NR and

thermoplastics were combined in the system due to some structure similarity, compatibility and phase interaction (Hanafi *et al.*, 2001). The main problem in composites blending reinforced with lignocellulosic fibre is the surface interaction between matrix and filler. Surface modification is very important in order to increase the interaction between hydrophobic polymers and the hydrophilic nature of lignocellulosic materials. Without chemical treatment, unstable interface interaction between polymer and fibre will be generated. This will create inefficient stress transferred from the matrix to the fibre and the beneficial reinforcement effect of the fibre remains underexploited (Chong *et al.*, 2010). Chemical or physical modifications on the surface of lignocellulosic particles were explored with the aim of changing the surface properties or creating reactive sites to improve the fibre-matrix interfacial bonding. By adding nanosized filler like GNP, composites' properties also will be enhanced (Shesmani *et al.*, 2013; Kang *et al.*, 2017).

Research on graphene has been aggressively explored since 2004 (Idumah and Hassan, 2016; Zakaria *et al.*, 2017). Graphene and its derivatives are added to the polymer matrix due to the ability to improve the physicochemical and mechanical properties of the materials (He *et al.*, 2017). Graphene is a one-layer carbon particle from the combination of  $sp^2$  hybridized linear carbon atoms that are aligned hexagonally in planar structure and very compact in the honeycomb crystal lattice structure. It also has a lot of potential as filler in nanocomposites (Geng *et al.*, 2009; Frasca *et al.*, 2015; Kumar *et al.*, 2017). The structure consists of a few layers of carbon atoms also known as graphene nanoplatlet (GNP) (Idumah and Hassan 2016). Graphene-based biocomposites also exhibit great barrier properties against gases, water vapour and ultra-violet light (Barra *et al.*, 2020). Thus, the hybridization of GNP in biocomposites will produce high barrier packaging materials.

The objective of this research was to determine the effectiveness of the hybridization of GNP with chemically modified STF to polymer matrix towards mechanical, physical and chemical properties. There are many types of research done on adding many types of fillers into polymer matrixes but studies on the effects of the hybridization of GNP and lignocellulosic fibres are still limited. The combination of microsize and nanosized fillers is expected to improve the overall performance of the materials obtained towards the application of high barrier food packaging materials.

## 2. Materials and methods

High-density polyethylene (HDPE) Etilenas

HD5301AA grade was obtained from Polyethylene (M) Sdn. Bhd. Natural rubber (NR) SMR-L grades were bought from Chemolab Supplies. For chemical treatment, hexadecyltrimetoxysilane (HDS) (Sigma Aldrich, >85%), ethanol (System, 95%), sodium hydroxide (NaOH) (analytical grade) and acetic acid (analytical grade) were bought from Iktikaf Resources Sdn. Bhd. (Malaysia). For antioxidant analysis, methanol (analytical grade), hydrochloric acid 6.0 N, gallic acid, Trolox, 2,2-diphenyl-picrylhydrazine (DPPH) and 2,4,6-Tris (2-picryl)-s-triazine (TPTZ) from Sigma (Germany), sodium carbonate anhydrous, iron (II) sulfate heptahydrate and sodium acetate 3-hydrate brand Bendosen, Folin ciocalteau reagent from Ajax Finechem, glacial acetic acid and iron (III) chloride hexahydrate from Merck (Germany) were used. GNP (grade KNG-150) with a thickness of 5-15 nm, diameter 5 µm and density of 2.25 g/cm<sup>3</sup> was supplied by KNANO.

### 2.1 Preparation of fibre

*Sansevieria trifasciata* trees, of ages more than 2 years were collected from a few areas in Klang Valley, Selangor, Malaysia. The leaves were cut and washed in order to remove dirt and soil. The leaves then were chopped using a slicer machine (Emura, Japan), dried using a drum dryer at 60°C for 5 hrs and ground to fine fibre. The fibre then was sieved to a size of 125 µm by using a sieve-shaker machine (Retsch, Germany).

### 2.2 Chemical treatment of *Sansevieria trifasciata* fibre

STF was pre-treated with 4% NaOH and stirred for 1 hr on a hot plate stirrer at 50°C before being neutralized with acetic acid. The sample then was treated with a 1% silane coupling agent (HDS) prepared in an ethanol mixture (80/20 v/v ethanol/water) by using ultrasonic equipment for 10 mins. After that, the mixture was stirred continuously for another 1 hr on a hot plate stirrer at a temperature of 60°C. The sample then was filtered and rinse with ethanol. The pH of the sample was maintained at 4. The sample was dried in a vacuum oven at 90°C until dry (3-5 hours of time).

### 2.3 Preparation of composites

Composite blending was done by using an internal mixer machine (Brabender W50EHT, Germany). Processing parameters used were 135°C for temperature and rotor speed at 55 rpm for 15 mins. NR/HDPE composition of 40/60 was used in the system while filler loading was 20%. HDPE was added to the internal mixer, followed by NR. After 5 mins, treated *Sansevieria trifasciata* (TSTF) and GNP were incorporated into the mixer to complete the blending process. Composites obtained were pressed in a hot press machine (Labtech, Sweeden) at 145°C to produce samples with 1 mm and 3

mm of thickness. The total pressing time was 12 mins.

### 2.4 Tensile test

Tensile tests were carried out according to ASTM D412 on an Instron Universal Testing Machine (model 5567, United Kingdom). Dumb-bell-shaped specimens of 1 mm thickness were cut from the hot press sheets by using a die cutter. A crosshead speed of 50 mm/min was used and the test was performed at 25±3°C. Seven specimens were used and the average was calculated respectively.

### 2.5 Water absorption test

The water absorption test was done according to ASTM D 570. Samples were pre-conditioned in an oven for 24 hrs at 50°C prior to soaking in water. The sample was weighed after 24 hrs of water soaking, after 3 days and repeated every week until a constant weight was achieved or samples failure was detected. The percentage of water absorption was calculated based on this equation:

$$\text{Percentage of water absorption} = \frac{M_w - M_d}{M_d} \times 100\%$$

Where  $M_w$  = Mass of wet sample and  $M_d$  = Mass of dry sample

### 2.6 Water contact angle test

Water contact angle test was done by using a contact angle meter (Holmarc HO-IAD-CAM-01A, India). A picture of the water droplet on a flat surface of the composites was captured and the degree of the contact angle was recorded. The contact angle of the materials must be greater than 90° for the classification of hydrophobic materials.

### 2.7 Morphological study

A morphological study was done by using a Field Emission Scanning Electron Microscopy (FESEM) Machine (Zeiss Supra 55VP, Germany). Samples of surface from treated fibre with NaOH-HDS were observed in this study. Samples were coated with platinum for the electron emission process.

### 2.8 Determination of total polyphenol content and antioxidant activity

#### 2.8.1 Extraction of samples

Samples of untreated *Sansevieria trifasciata* fibre (USTF) and treated *Sansevieria trifasciata* (TSTF) underwent extraction with distilled water at a ratio of 24 mg to 10 mL (w/v). Each mixture was placed in a centrifuge tube and vortex for 1 min and agitated at 150 rpm at room temperature in a shaker for 1 hr. The mixture was then centrifuged at 8,500 RPM for 10 mins

using Biofuge Primo (Heraeus). The supernatant was filtered through Whatman No. 541 filter paper (Merck) to obtain a clear extract. The filtrates were assayed for their total phenolic content (TPC) and antioxidant activity assay (AOA), as described below. All experiments were run in triplicate.

### 2.8.2 Determination of total phenolic content

Total phenolic in all samples was determined with Folin–Ciocalteu assay (Singleton and Lamuela-Raventos, 1999) by using gallic acid as a standard phenolic compound. The fresh weight of each sample was converted into dry weights on the basis of the moisture content. Briefly, 1 g of sample was dissolved in 20 mL of distilled water, then 50  $\mu$ L of appropriately extracts solutions and standard gallic acid solutions (12.5, 25.0, 50, 100, and 200  $\mu$ g/mL) were mixed with 50  $\mu$ L of distilled water in a well of 96-well plate, then 100  $\mu$ L of Folin-Ciocalteu reagent solution (prediluted 10-fold with distilled water) was added. After 6 mins, 100  $\mu$ L of 7.5% (w/v)  $\text{Na}_2\text{CO}_3$  was added and mixed gently. The reaction mixture was kept in dark for 2 hrs and its absorbance was measured at 765 nm against distilled water as a blank solution using the microplate reader. The total phenolic content (TPC) was expressed as mg gallic acid equivalents (mg GAE/g dry weight), which was determined from known concentrations of gallic acid standard. Data were reported as a mean  $\pm$  standard deviation for three replications.

### 2.8.3 Determination of antioxidant activity

#### 2.8.3.1 Determination of free radical scavenging activity

The hydrogen atom or electron donation ability of the corresponding extracts and some pure compounds was measured from the bleaching of purple-coloured methanol solution of 2,2-diphenyl-1-picrylhydrazyl (DPPH). This spectrophotometric assay uses stable radical DPPH as a reagent, according to a slightly modified method of Lu and Yeap Foo (2000). A volume of 100  $\mu$ L of the extracts was added to 200  $\mu$ L of a 0.007% methanol solution of DPPH. After a 40 min incubation period at room temperature, the absorbance was read against a blank at 517 nm using a microplate reader. The percentage of inhibition of free radical DPPH by the extracts was calculated as followed:

$$\text{Inhibition (\%)} = (A_{\text{blank}} - A_{\text{sample}} / A_{\text{blank}}) \times 100$$

Where  $A_{\text{blank}}$  is the absorbance of the control reaction (containing all reagents except the test compound), and  $A_{\text{sample}}$  is the absorbance of the test compound.

#### 2.8.3.2 Ferric reducing antioxidant power assay

The ferric reducing antioxidant power (FRAP) assay was performed as previously described by Benzie and Strain (1996). This method was developed to measure the ferric reduction ability of plasma at a low pH. When the ferric 2,4,6-tripyridyl-s-triazine complex ( $\text{Fe}^{3+}$ -TPTZ) is reduced to the ferrous form ( $\text{Fe}^{2+}$ -TPTZ), the intense blue colour is developed. Briefly, The FRAP reagent was prepared by mixing 10 volumes of 300 mM acetate buffer (pH 3.6), with one volume of 10 mM TPTZ in 40 mM HCl and with one volume of 20 mM  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$  and the mixture was then incubated at 37°C for at least 10 mins. A total of 20  $\mu$ L of extract solution and 80  $\mu$ L of distilled water were added to 200  $\mu$ L of freshly prepared FRAP reagent in a well of 96-well plate. After four min, the absorbance was read using a microplate reader at 593 nm against the reagent blank, which was prepared by the same procedure described above except that the extract solution was substituted by 20  $\mu$ L of water. The FRAP value was calculated and expressed as milligram of Trolox equivalents per 100 g of dry weight sample (mg TE /100 g dw) based on a calibration curve plotted using Trolox as a standard at a concentration ranging from 12.5 to 200  $\mu$ g/mL.

### 2.9 Spectrophotometric measurements

All spectrophotometric measurements were taken using BIOTEK GEN5 Eon Microplate Spectrophotometer (Winooski, Vermont, USA) in a 96 well plate.

### 2.10 Statistical analysis

Each of the measurements described above was conducted in triplicate and the mean  $\pm$  SD (standard deviation) was reported. The data collected were statistically analysed using the Statistical Analysis Software (SAS) package (version 9.4 of SAS Institute, Inc. Cary, NC, 2008). Statistically significant differences ( $p < 0.05$ ) in the antioxidant properties of the samples were determined by one-way analysis of variance (ANOVA). Duncan Multiple Range Test (DMRT) was used to determine significant differences between the means.

## 3. Results and discussion

### 3.1 Chemical modification

Using treated STF has produced tremendous effects on almost all of the properties investigated. There were four steps involved in the chemical modification of STF that improved the interaction which were hydrolysis, self-condensation, adsorption and grafting (Xie *et al.*, 2010). In hydrolysis, silane monomers were hydrolysed in the



presence of water and catalyst (NaOH) liberating alcohol and reactive silanol groups will yield. After that, self-condensation will happen. This step will leave free silanols to be absorbed by the hydroxyl groups in the natural fibres. The condensation rate of silanols was controllable by adjusting the pH of the hydrolysis system. An acidic pH environment was preferable to accelerate the hydrolysis rate of silane but slow down the condensation rate of silanols. In this research, pH 4 was the optimum for the best rate of the condensation process. Then adsorption process will occur where the reactive silanol monomers or oligomers were physically adsorbed to hydroxyl groups of STF by hydrogen bonds on the fibre surface and/or cell walls (depending on the molecular size of silanol monomer/oligomers formed). The last process was grafting. Under heating conditions, the hydrogen bonds between the silanols and the hydroxyl groups of fibres can be converted into covalent bond (-Si-O-C-) bonds and liberate water (Xie *et al.*, 2010).

### 3.2 Tensile test

Figure 2 shows the value of tensile strength of untreated and treated composites with different percentages of GNP loading. Pre-treatment with NaOH has produced a rougher surface because of the lignin, hemicellulose, pectin, wax and impurities removal. This will increase the surface interaction between the matrix and fibre. The hydrophobic surface of the polymer matrix and the hydrophilic surface of the fibre that has been modified with silane coupling agent will give better filler dispersion than the untreated composites. Stronger interaction between filler and matrix will facilitate a more efficient energy transfer process and a tougher composite will be produced (Chong *et al.*, 2010). Fibre dispersion has been identified as a major factor influencing the properties of short fibre composites and a major challenge in natural fibre composites (Pickering *et al.*, 2016).

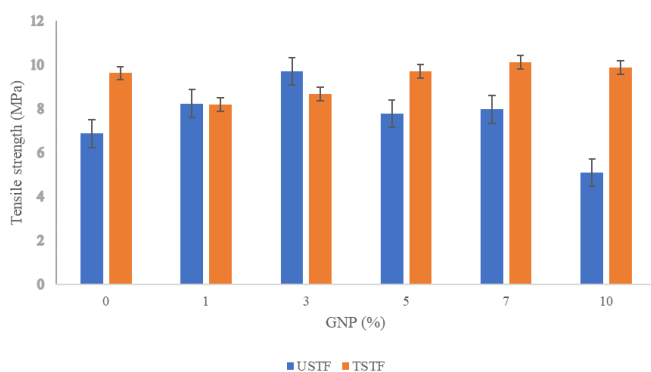


Figure 2. Tensile strength of untreated and treated composites with different percentages of GNP loading

The addition of GNP in the composite system

increased the tensile strength of treated samples compared to the untreated. In the untreated system, GNP loading between 1-3% was able to increase the value of tensile strength due to the synergistic effect of GNP. However, after 5% of GNP, the composite's tensile strength will decrease caused of the agglomeration of the particle that led to poor filler–matrix interaction. Graphene is a nano-size filler that has the tendency to form agglomeration after the critical point of loading due to its internal molecular force such as  $\pi$ - $\pi$  and Van der Waals force (Yaragalla *et al.*, 2015). The addition of 1-3% GNP in the treated composites decreased the tensile strength. A small amount of GNP had given a dilution effect on the composite system. After GNP loading increased (more than 5%), the composites system become more stable due to better interaction of each component. The rougher surface of TSTF with silane contributed to more mechanical interlocking of the materials which led to better strength. Results showed that the chemically modified STF has improved GNP distribution and dispersion in the composite system. Treatment is done also increased the GNP critical point of loading in the hybridized composites.

Figure 3 shows the tensile modulus of untreated and treated composites with different percentages of GNP loading. Overall, tensile modulus will increase as filler loading increases because the amount of filler will contribute to material rigidity and stiffness. The tensile modulus for untreated STF is lower than the treated composites. Treatment done has produced smaller particle size with rougher surface materials and increased the value of modulus due to better interaction of filler-matrix that has produced stiffer materials. The more rigid the materials, the more energy will be needed to elongate them. Research by Siakeng *et al.* (2020) also showed that treated lignocellulosic fibre has enhanced the tensile modulus of the hybrid composites obtained.

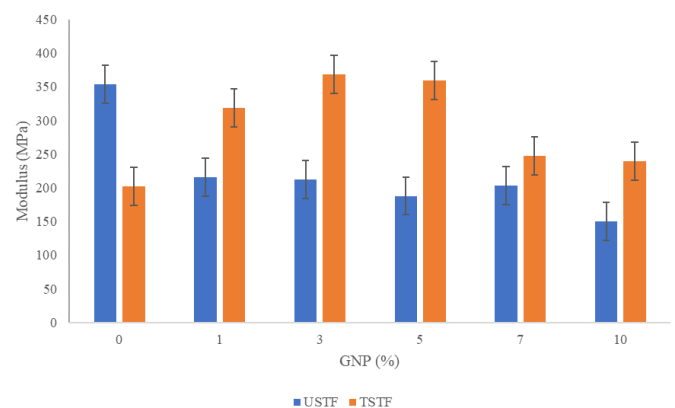


Figure 3. Tensile modulus of untreated and treated composites with different percentages of GNP loading

Generally, the addition of nano-sized filler will increase tensile strength and composite modulus but filler loading, types of filler, filler dispersion and

interaction between filler-filler or filler-matrix will also give a big impact on the overall performance of the lignocellulosic-based hybrid composites. Poor wettability, agglomeration, nanofiller dispersion and distribution will be improved when the chemically modified lignocellulosic fibre is used in the hybrid composites' system (Siakeng *et al.*, 2020).

### 3.3 Water absorption test

Water absorption tests were performed for pure HDPE, matrix NR/HDPE (40/60), and composite samples with and without GNP either treated or untreated to observe the level of water absorption for each material. A certain period of time has been studied in this research, i.e 24 hrs, day 3 and every week until a constant weight was achieved. Figure 4 shows the results of water absorption of each material (i.e. HDPE, matrix (NR/HDPE), untreated and treated composites with/out GNP). The result showed that untreated composites (with and without GNP) have given a higher amount of water absorption, followed by treated composites (with and without GNP). Composites that contain natural fibre without modification usually will absorb water due to numerous reasons such as cellulose and hemicellulose content, humidity, hydrophilic nature of the fibre, presence of voids, temperature and viscosity of the matrix (Jawaid and Khalil, 2011). Samples with pure HDPE and matrix (NR/HDPE) have given minimum water absorption because both samples are originally hydrophobic. The incorporation of STF has contributed to a higher amount of water absorption in the composite's system but the modification of fibre by using silane-coupling agent has improved the interaction. Good interaction produced fewer microvoids in the composites due to better STF-GNP interaction with the matrix. Fibre treatments usually fill the surface's pores and create a barrier for water molecules to enter into fibre (Akas *et al.*, 2017).

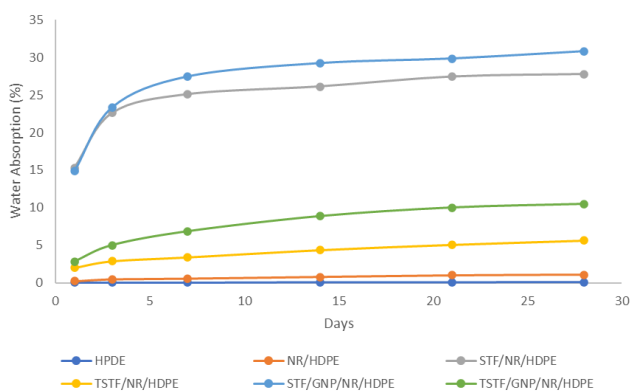


Figure 4. Water absorption of HDPE, matrix (NR/HDPE), untreated and treated composites with/out GNP

### 3.4 Water contact angle test

Figure 5 shows the contact angle value of untreated

and treated composites with different percentages of GNP loading. Lignocellulosic materials are very sensitive to moisture and also hygroscopic. This test was done to observe the hydrophobicity surface of materials either treated or untreated. Generally, if the water contact angle is smaller than  $90^\circ$ , the solid surface is considered hydrophilic and if the water contact angle is larger than  $90^\circ$ , the solid surface is considered hydrophobic (Förch *et al.*, 2009). Many polymers exhibit hydrophobic surfaces. Contact angles are very sensitive to contamination. According to Fowkes (1964), if the liquid molecules are strongly attracted to the solid molecules, then the liquid drop will completely spread out on the solid surface, corresponding to a contact angle of  $0^\circ$ . This is often the case for water on bare metallic or ceramic surfaces. However, the presence of contaminants on the solid surface can significantly increase the contact angle.

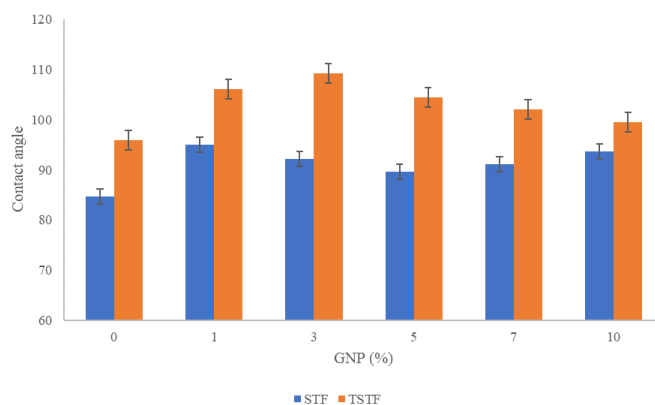


Figure 5. Contact angle of untreated and treated composites with different percentages of GNP loading

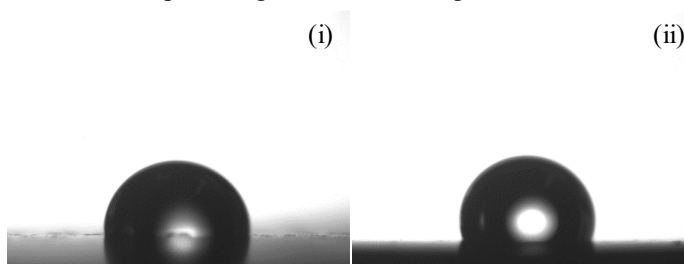


Figure 6. Picture of water droplets from contact angle test, (i) untreated and (ii) treated composites

Figure 6 shows the picture of water droplets from the contact angle test for treated and untreated composites captured by the apparatus. All contact angle values for treated composites are greater than  $90^\circ$ . This is the indicator for a hydrophobic surface of the materials obtained. Treated composites had given higher value of contact angle ( $95.96^\circ$ ) compared to the untreated composites ( $<90^\circ$ ). The addition of GNP contributed to producing more hydrophobic surfaces. 3% of GNP loading had given the highest value of contact angle.

### 3.5 Morphological study

Figure 7 shows the FESEM micrograph for treated composites with different percentages of GNP loading at

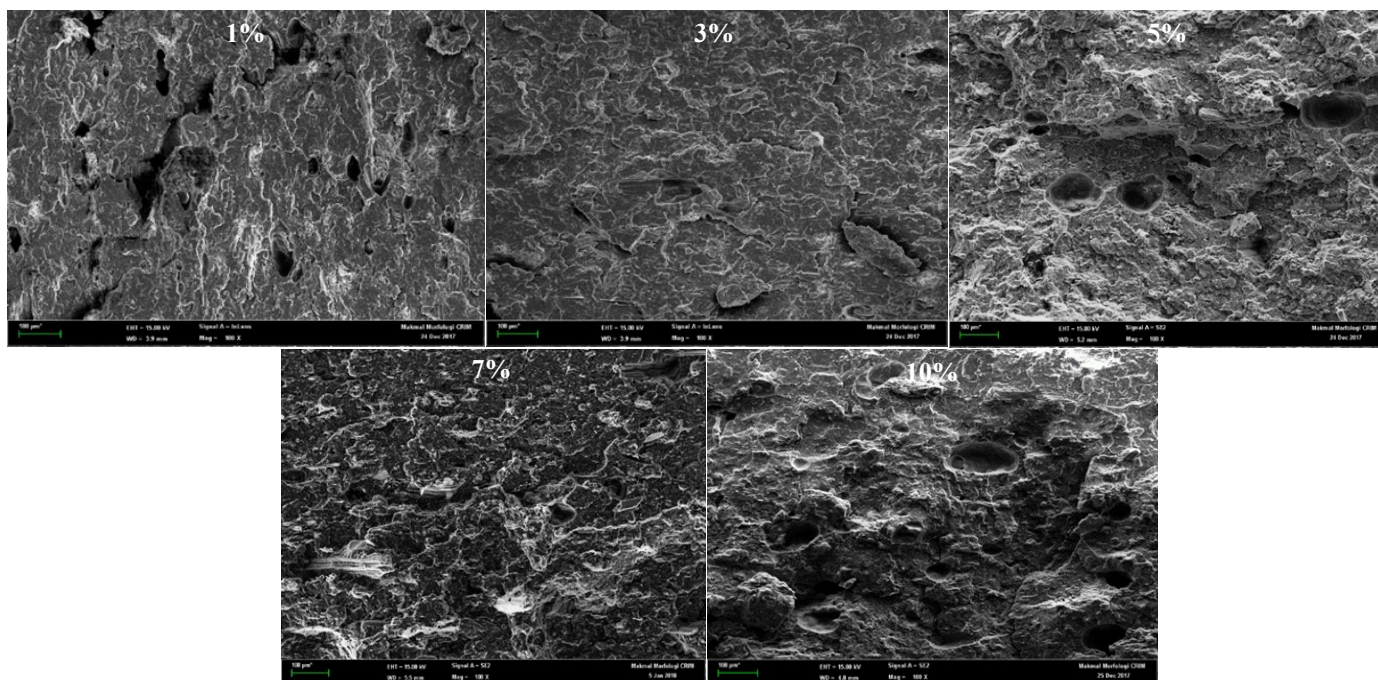


Figure 7. FESEM micrograph for treated composites with different percentages of GNP loading (1-10%) at 100× magnification. This scale was chosen to observe the dispersion of filler in the matrix. The 1% GNP content in the composite has produced more microvoids compared to another amount. The addition of GNP had shown less particle agglomeration for a composition of 3-7% GNP. From micrograph of the sample with a GNP content of 3% has shown the best particle dispersion and distribution compared to other compositions. This explained why most of the results characterized have favoured the composition of 3% GNP. This good interaction is also reflected in the matrix in the composite. The matrix in 3% GNP composition has shown a softer look and more uniform.

Less fibre pull-out was observed in most of the micrographs. This indicated that better interaction was achieved after the chemical modification with silane. Many fibre pull-outs showed that the bonding/interaction between fibre and matrix is weak. While cracks on the matrix surface showed either fibre loading is not enough or fibre distribution in the composite is heterogeneous (Jawaid *et al.*, 2013).

### 3.6 Antioxidant activities

Results in Table 1 showed that treatment done by using NaOH and silane had decreased the total phenolic content of STF while free radical scavenging activity and ferric reducing antioxidant power (FRAP) had increased with the treatment. Pre-treatment of lignocellulosic

biomass changes the chemical composition and alters the surface conditions of fibres, which may improve composite performance (Rahman *et al.*, 2017). Pre-treatment with NaOH has removed the phenolic content of STF. Antioxidant activity in treated fibre was also higher than in untreated fibre, due to the removal of the impurities during the treatment. This has proven the efficiency of the treatment done on fibre. Alkaline pre-treatment of STF by sodium hydroxide has been shown capable of reducing lignin content, enhancing digestibility and hydrolysis towards higher antioxidant activities (Ibrahim *et al.*, 2013). This will produce packaging materials with natural antioxidant properties to prolong the shelf-life of food products.

## 4. Conclusion

Blending of GNP with chemically modified STF/NR/HDPE together has produced homogenous composites. Adding NR to the composite system has produced less rigid and softer surface composites. Chemical modification by using silane-coupling agent has given better results for most of the characterization done. The amount of 3% GNP loading has given the best result for tensile modulus, water contact angle and also for morphological study. Composites obtained have the potentials to be applied as packaging films to prolong the shelf life of food products due to their high barrier properties to water and moisture. The value is also higher

Table 1. Results for antioxidant analysis

Samples	TPC (mg GAE/ 100 mL)	FRAP (mg TE/ 100 mL)	DPPH (% inhibition at 2.4 mg/mL)
TSTF	8.38±0.06 <sup>b</sup>	37.47±0.48 <sup>a</sup>	37.79±1.03 <sup>a</sup>
USTF	15.16±0.12 <sup>a</sup>	24.87±0.16 <sup>b</sup>	35.43±0.34 <sup>b</sup>

Values are presented as mean±SD. Values with different superscript within the same column are significantly different ( $p < 0.05$ ).



than typical packaging plastic (i.e HDPE). Materials obtained are also suitable for green polymer applications, natural antioxidant packaging films and biodegradable packaging applications.

### Conflict of interest

The authors declare no conflict of interest.

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