The effect of stearic acid and different loading treated and untreated EFB fiber on the tensile, structural and chemical properties of polypropylene/recycled acrylonitrile butadiene rubber/empty fruit bunch composites

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Abstract. A new developed of polymer materials made up of polypropylene (PP), recycled nitrile gloves (NBRr) and empty fruit bunch (EFB) fiber was fabricated. Recycled nitrile glove was blended together with PP plastic to reduce the use of petroleum based and to solve the issue of waste discarded at the landfill. EFB fiber was incorporated to lower the production cost of composite. Due to incompatibility of different phases, stearic acid treatment was used to improve the EFB fiber polarity. Thus, the effect of different EFB loading for untreated and treated stearic acid of PP/NBRr/EFB composite was studied. In this work, all the materials were mixed well using a heated two roll mill with temperature 180 °C within 9 minutes. The EFB loading were varied from 0 to 30 phr. The analysis of tensile properties was tested followed the ASTM D638, Type IV, testing procedure, to analyze tensile strength, Young’s modulus and elongation at break. The structural properties of the fracture sample surfaces were observed using Scanning Electron Microscope (SEM) and the chemical properties was analyzed using Fourier Transform Infra-Red (FTIR). Composites with stearic acid treatment have shown higher tensile strength compared to untreated EFB fiber. Besides, the micrograph structure surface from scanning electron microscopy analysis showed better fiber and matrix interaction, as the treated-EFB fiber is well encapsulated with the PP matrix. From the FTIR analysis, the intensity peak of OH for PP/NBRr/EFB/SA was reduced due to the removal O-H bond of cellulose from fibers during the surface modification of fibers.

1. Introduction

The concern of environmental preservation has been strongly emphasized. Reducing the consumption of plastics is one of the ways to minimize the harmful effects on the environment. Many research has interest to develop new polymer materials using natural fibers such as jute, sisal, pineapple, straws of rice, sugarcane bagasse, oil palm frond, empty fruit bunch [1], etc. Natural fibres would produced low cost of production compared to synthetic fiber, low density and have high specific properties [2].
replacement of natural fibers will reduce the amount of synthetic waste discarded at the landfill. Natural fibers act as fillers in polymer matrix composites. Polypropylene (PP) is one type of thermoplastic used as a matrix. It also has low density, high mobility and excellent abrasion resistance.

Nitrile rubber is a synthetic rubber made up of copolymer of acrylonitrile and butadiene. Highly resistant to oil and acid [3] as well as its superior strength results in high demand for nitrile glove production. It was widely used in medical purposes due to non-allergic to consumers and high resistance to puncture. Despite that, abundant nitrile gloves have been discarded since it was applied for single use only, and cannot be disposed and degraded. Therefore, the recycled nitrile gloves will blend with polypropylene and empty fruit bunch fiber to produce new thermoplastic elastomer composites.

However, the incompatibility issue between natural fiber and polymer matrix results in difficulty to make achieve a strong interfacial adhesion [4]. To improve the incompatibility between natural fibers and the matrix, coupling agents are essentially use. Coupling agents such as silanes, benzoyl peroxide, isocyanate [1] are widely used. The costs of those coupling agent are quite high and their application required a complex procedure [5]. Thus, the use stearic acid (SA) which acts as dispersing agent is more practical and cheaper. SA contains both a carboxyl group which can form hydrogen bonds with hydroxyl groups of cellulose and a hydrocarbon chain which is compatible with hydrophobic groups of polymer matrix. Previous study reported that the use of stearic acid can improve the fiber dispersion in polymer matrix [6].

In this study, polypropylene (PP) composites fabricated with recycled nitrile glove and empty fruit bunch (EFB) fibers were produced. Tensile, structural and chemical properties were investigated and compared between the stearic acid treated and untreated EFB fiber composites.

2. Experimental

2.1. Materials
Isotactic polypropylene (PP) (code 6331: density 0.9g/cm³) was supplied from Titan PP Polymers Sdn Bhd. The recycled acrylonitrile butadiene rubber (NBRr) was obtained from Juara One Resources (M) Sdn Bhd. Empty fruit bunch (EFB) was collected at United Oil Palm Industries Sdn. Bhd.

2.2. Material Preparation, Mixing and Moulding
Recycled acrylonitrile butadiene rubber (NBRr) was ground and sieved to size of 150-300µm. Empty fruit bunch (EFB) fibre was also cut and grind into a smaller size. Prior to pretreatment using sodium hydroxide (NaOH) solution, EFB fibre was dried in an oven for 24 hours at 80 °C. 100g of EFB fibre was soaked and stirred in NaOH solution for one hour and rinsed with distilled water. EFB fibre was then soaked in stearic acid (SA) solution for one hour. EFB fiber was dried in the oven for 24 hour at 80 °C. All materials were weighted followed the formulation given in Table 1. The samples have to be dried at 80°C for 24 hour prior mixing. A heated two roll mill machine from Fang Yuan Instrument (DG) Co. Ltd was used to melts mixed all the substances at 180 °C with rotor speed at 15 rpm.

| Materials | Composites Loading |
|-----------|--------------------|
| PP        | 70                 |
| NBRr      | 30                 |
| EFB       | 0                  |

An electrical heated hydraulic press model GT-7014-A30C with force 2500 kg/cm³ was used to compress the mixing compounding. This hot press machine was heated up to 180 °C for seven minutes preheating, two minutes compression and two minutes of cooling down. Then, the thin sheet sample was cut into dumbbell shape by using a Wallace die cutter model S6/1/6.A.
2.3. Testing and Characterization Analysis

2.3.1. Tensile Testing. A Universal Testing Machine (UTM) model Instron 3366 was used to conduct the tensile test. All the samples were precisely measured the length, width and thickness, using a vernier caliper. The tensile test standard method used is ASTM D 638, Type IV, with a dimension of width, 6 mm, length 140 mm and thickness 1mm. The initial jaw separation distance of the tensile tester is 50 mm and the testing is carried out at a cross head speed of 5 mm/min. The tensile tests were repeated five times for each series of formulation.

2.3.2. Scanning Electron Microscope (SEM). Studies on the structural morphology of the tensile fractured composites were done by using a Scanning Electron Microscope (SEM) model Zeiss SUPRA 35VP. The selected fracture specimen was coated with a thin platinum layer using Auto Fine Coater JFC1600 to avoid electrostatic charging and poor resolution.

2.3.3. Fourier Transform Infra-Red (FTIR). Fourier Transform Infra-Red (FTIR) analysis of each material and composites was evaluated using FTIR spectrometer (Perkin Elmer Series 2 spectrometer). FTIR spectra were recorded by plotting the wavelength against transmittance. The spectrum resolution is 4 cm\(^{-1}\) and the scanning range is from 550 to 4000 cm\(^{-1}\).

3. Result and Discussion

3.1. Tensile Properties

Tensile strength of PP/NBRr/EFB fibers composites with and without stearic acid (SA) treatment was illustrated in figure 1. The tensile strength of untreated composites was decreases from 17.14 MPa to 12.5 MPa, as the EFB fiber loading increased from 5 to 30 phr. The decreasing of the tensile strength was due to poor dispersion of the filler in the matrix, which increase the interfacial defects or debonding between the filler and matrix [7]. At 30 phr fiber loading, the highly fiber content will lead to a strong inter filler hydrogen bonding which allows them to cling together, and thus, resist the dispersion of the filler. The weak interfacial bonding leads to reduce the tensile strength of the composites [8]. However, for the treated-EFB fiber composites, the tensile strength showed an improvement about 4.1 – 5.8 %. It was due to the existence of the carboxyl group of the stearic acid, which able to form hydrogen bonds with hydroxyl group of cellulose (hydrophilic group) and a hydrocarbon chain with polymer matrix (hydrophobic group). These can improve the mechanical properties of the composites by improving the chemical adhesion between the matrix and the filler [5].

![Figure 1. Tensile strength of PP/NBRr/EFB composites](image)

The increasing of EFB fiber loading resulted in enhancing the Young’s modulus since it represents the stiffness of the composites. Figure 2 shows the Young’s Modulus of PP/NBRr/EFB fibers composites with and without stearic acid treatment. Based on the result obtained, the Young’s Modulus of
untreated composites was found to be increased from 602 MPa to 881 MPa. The modulus was enhanced due to the fibrous form of EFB fiber that may carrying more tensile load and contain much stiffer than polymer matrix. Thus, adding filler capable of enhancing composite stiffness in which resists the deformation of composites. Nevertheless, the Young’s Modulus for stearic acid treated- EFB fibers was improved about 3 – 13.8 % higher compared to the untreated EFB fibers composites for similar filler content. Treated fibers reinforce polymer behave stiffer and could withstand higher stress at the same strain portion. This was due to the improvement of better adhesion between fiber and matrix by the chemical interaction.

Figure 2. Young’s modulus of PP/NBRt/EFB composites

Figure 3 below shows the result for elongation at break of the PP/NBRt/EFB composites with and without stearic acid. The elongation at break, Eb, of untreated composites was decreased from 9.1 % to 5.8 %, as the fiber loading increased. The composites become stiffer and harder when the fiber loading in composites increased. However, the elongation at break, Eb of the treated composites was 8.4 % to 5.1 %. The lower Eb might due to the improved adhesion in the presence of dispersing agent. The restricted of mobility of polymer segment resulted to reduce the composites elongation. Stearic acid treatment had improved the interfacial adhesion between fibre and rubber matrix, produced the composites with a better strength and stiffness.

Figure 3. Elongation at break PP/NBRt/EFB composites
3.2. Morphological Studies.

Figure 4 (a) and (b) show the micrographs of raw untreated EFB fiber and stearic acid treated EFB fiber, respectively. The surface of untreated EFB fiber seems smoother while the treated EFB fiber has rough surface due to the fibrillation during the pre-treatment. Figure 4 (c) and (d) observed the SEM micrograph of the fracture surface of treated PP/NBRr/EFB composites and untreated PP/NBRr/EFB composites at 70/30/5. The addition of EFB filler into PP/NBRr matrices improved the rigidity of composite hence change the composites from ductile to even more brittle.

Figure 4. SEM micrograph of raw EFB fiber and untreated PP/NBRr/EFB composites; a) untreated EFB, b) SA-treated EFB, c) 70/30/5-untreated and d) 70/30/5-treated

Figure 5 (a) and (c) shows the micrograph of the rupture surface of untreated PP/NBRr/EFB composites with different fiber loading. From the observation, there were many holes present, due to the pull out of EFB fiber and NBRr from the PP matrix. Besides, EFB filler detachment site was observed due to weak bonding with PP/NBRr matrices. Figure 5 (b) and (d) shows the SEM micrographs of treated PP/NBRr/EFB composites. 70/30/15-treated composites (Figure 5b) showed less holes and gap presence. It clearly has shown that, more EFB fibre was encapsulated on the PP matrix. It can be seen that good with less EFB detachment sites was observed for treated composites. This could provide a better tensile strength of PP/NBRr/EFB composites. However, at higher EFB loading, poor adhesion between filler and matrix was observed with presents of voids and holes.
3.3. **Fourier Transform Infra-red (FTIR) Analysis**

Figure 6 shows FTIR spectra comparison of EFB, treated-EFB, NBRr, PP/NBRr, PP/NBRr/EFB and treated PP/NBRr/EFB composites in the region of 500 to 4000 cm\(^{-1}\). The C-H stretching group arise at peak 3000 to 2894 cm\(^{-1}\) for raw EFB and treated EFB. The intensity of the C-H group was higher after stearic acid (SA) treatment. It is because of the interaction of the lignin with the SA to produce a C-H bond. Natural fibers are fiber-reinforced by cellulose fibre, since they consist of microfibrils in an amorphous lignin and hemicellulose matrix. Besides, there also a C=C stretching group of peak between 1670 to 1600 cm\(^{-1}\) at raw EFB and treated-EFB is due to the lignin in the fibers.

The O-H stretching group at peak 4000 to 3000 cm\(^{-1}\) only arises at PP/NBRr/EFB-treated composite. It was due to the removal O-H bond of cellulose from fibers during the surface modification of fibers. The presence of the C=O stretching group at a peak between 2000 to 1650 cm\(^{-1}\) was due to the interaction between the SA molecules with the EFB fibers. Thus, it also shows the good adhesion between the EFB filler and PP/NBRr matrices and on other hand reduces the incompatibility problem of the composite. As a conclusion, surface modifications with SA treatment on EFB are able to improve the incompatibility of the composites.

**Figure 5.** SEM micrograph of PP/NBRr/EFB composites with and without treatment; a) 70/30/15-untreated, b) 70/30/15-treated, c) 70/30/30-untreated and d) 70/30/30-treated

EFB detachment
Figure 6. FTIR spectra of raw EFB fiber, treated-EFB fiber, raw NBRr, PP/NBRr/EFB composite and PP/NBRr/EFB/SA composite

4. Conclusion
This work presented a way to improve the fiber-matrix incompatibility which is the main disadvantages of thermoplastic reinforced natural fibres. The treatment using stearic acid can act as coupling agent by improving the tensile properties of PP/NBRr/EFB composites. The tensile strength and elongation at break of PP/NBRr/EFB composites was decreasing and young’s modulus was increased as the EFB fiber loading increase. However, stearic acid treatment resulted to better tensile strength which ranges from 13.4 MPa to 18 MPa. Besides, the SEM micrograph showed an improved of interaction between EFB fibre and PP/NBRr matrix with stearic acid treatment.

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