

Material Treatment of Polypropylene (PP) / Recycle Acrylonitrile Butadiene Rubber (NBrr) and Banana Skin Powder (BSP) using Thermal Mixing Techniques

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ABSTRACT

One of the most significant current discussions in material properties is the chemical treatment effect on the proposed materials. In this paper, the effects of NaOH and α -APS silane treatments on the characteristics, thermal and mechanical properties of polypropylene (PP)/recycled acrylonitrile butadiene (NBRr)/ banana skin powder (BSP) experimented. The thermal mixing process by heated two roll mills was used to melt mix the composites at a temperature of 180°C to produce six different fiber loading (70/30/5, 70/30/10, 70/30/15, 70/30/20, 70/30/25 and 70/30/30w/w). The effects of NaOH and α -APS silane treatment on PP/NBRr/BSP composites were analyzed using Tensile Test, Scanning Electron Microscopy (SEM), Water Uptake and Fourier Transform Infrared Spectroscopy (FTIR). The results showed that α -APS silane treated PP/NBRr/BSP composites has better improvement on mechanical properties when compared to the NaOH treated PP/NBRr/BSP composites, followed by the weakest mechanical properties of untreated PP/NBRr/BSP composites. These were due to good compatibility and stronger interaction between Si-O-Si moieties with the surface of fiber for α -APS silane treated BSP. The mechanical properties of PP/NBRr/BSP composites improve with decreasing BSP filler loading. This was due to better interfacial adhesion between both α -APS silane and NaOH treated BSP filler with PP/NBRr matrices while decreasing BSP filler loading. Furthermore, the chemical interactions in the composite before and after treatments were also investigated.

Keywords:

banana skin fiber, alkaline treatment, silane treatment, polypropylene, composites

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1. Introduction

Polyolefins are the major industrial polymers which are based on low-priced petrochemicals or natural gas and the required monomers are created by cracking or refining crude oil [1,2]. Low cost and good in mechanical and thermal properties are two of the reasons that Polypropylene (PP) is

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widely used. Besides, PP also has high crystalline as well because of the rigid polymer structure. These advantages can help in forming and toughness and high mechanical and thermal resistance to composite [3-6].

The Recycle Acrylonitrile Butadiene (NBR) can be seen widely used in industries for oil requirement and chemical resistance such as conveyor belting, hydraulic hose, roll covers, oil field packers and seals for all kinds of plumbing and appliance application [7-10]. The Nitrile rubber glove is one of an example product of NBR. It consists of monomer acrylonitrile, butadiene, and carboxylic acid and forms a synthetic polymer together. Plastics which modified by using NBR have higher tensile strength and flexibility. Thus, nitrile rubber gloves should be recycled and its natural fillers can be added to become cheaper and lighter composite materials [11,12]. Natural fiber such as banana fibers has following advantages: low density, inexpensive, harmless to human and environment, large in quantity, cannot deplete when used, high stiffness and strength properties, and finally, it is easily decomposed by living organisms [13,14]. Banana Skin Powder (BSP) had been chosen to be used on Polypropylene (PP) /Recycled Acrylonitrile Butadiene Rubber (NBRr) composite in order to minimize waste production as well.

There are many studies carried out by researchers on polymer composites to improve their mechanical and thermal properties by reinforcing with natural fibers, such as High Density Polyethylene/ Banana Peel Fibers (HDPE/BP) [12], Banana Fiber/ Polyester, Banana Fiber/ Epoxy, and Risk Husk Powder filled Polypropylene/ Recycled Acrylonitrile Butadiene Rubber (PP/NBRr/RHP)[8]. Therefore, due to the hydrophobic effect between the hydrophilic fiber and hydrophobic matrices, many studies on chemical modification or treatment had been carried out to remove the hydroxyl groups from lignin and cellulose of natural fiber to improve strength and fitness of fiber as well as adhesion between matrices and fiber in natural fiber composites [15].

In this paper, a novel experiment based on the effect of NaOH and α -APS silane treatment in the PP/NBRr/BSP composites was investigated. The effects of NaOH and α -APS silane on the thermal and morphological properties were analyzed. The primary goal of this study is to improve the materials characterization based on analysis such as Tensile Test, Scanning Electron Microscopy (SEM), Water Uptake and Fourier Transform Infrared Spectroscopy (FTIR). The rest of the paper is organized as follows: Section 2 explains the methodology of the experiment. Section 3 describes the result and discussion and finally, section 4 concludes this work.

2. Experimental

2.1 Materials

PP Grade 6331 with a density of 0.9g/cm^3 and a melt flow index of 1.4g/min at 230°C was obtained from Titan Pro Polymers (M) Sdn. Bhd. Johor, Malaysia. Recycled Acrylonitrile Butadiene Rubber (NBRr) powders with 33% acrylonitrile content were processed from the used nitrile gloves obtained from Juara One Resources Sdn. Bhd. Penang, Malaysia. Firstly, the nitrile gloves were mesticulated by using two roll mills machine. Next, the NBRr were grinded and sieved to obtain the particle size of $150\ \mu\text{m}$. A coupling agent α - aminopropyltrimethoxysilane (APS) was provided by Alfa Aesar (M) Sdn Bhd, Malaysia and NaOH pellets were bought from AR Alatan Sains (K) Sdn Bhd, Malaysia. Banana skins were collected from a fried banana hawker stall near Jejawi Perlis, Malaysia. The banana type used is *Musa paradisiaca* var. Awak or also known as 'Pisang Awak'[16]. The banana skins were cut into smaller pieces with a pair of scissors. Then, they were dried at 80°C for 24 hours in an oven. The dried banana skins were grinded, followed by sieving the banana skin powder to a particle size of $150\text{-}300\ \mu\text{m}$.

2.2 Formulations

The formulations of PP/NBRr/BSP components are as presented in Table 1, PP/NBRr/BSP composites with and without 5 w/w of NaOH and 5 w/w of α -APS silane treatment.

Table 1

Formulations of BSP filled PP/NBRr Biocomposite with and without NaOH and α -APS silane

Material	Amount (w/w)					
PP	70	70	70	70	70	70
NBRr	30	30	30	30	30	30
BSP	5	10	15	20	25	30

2.3 Preparation of Composites

Formulations of BSP filled PP/NBRr biocomposites were shown in Table 1. Before compounding, the PP granule, NBRr powder and BSP powder were dried by using a vacuum oven at 80°C for 24 hours. In order to mix the composite, the heated two roll mills were used with a temperature of 180°C. PP was allowed to melt for 4 minutes before adding NBRr matrices for another 3 minutes into the mixer. Then, BSP was added into the mixture for the fabrication of this biocomposite after 7 minutes of mixing. Another 3 minutes were required for the mixing in order to obtain the stabilization torque. The whole mixing process was taken 10 minutes for all samples. 1mm sheet of test samples was created by allowing the composites undergoing a compression-molded process by using Go-Tech compression moulding machine type GT-7014-A30C, manufactured by Go-Tech Testing Machines Inc, Taichung, Taiwan. Next, the test samples composites were undergoing preheating for 7 minutes at 180°C, followed by compression at 1000psi for 3 minutes, and then cooled for another 3 minutes. After the moulding process, a Wallace die cutter model: S6/1/6.A was used to cut the moulded samples to dumbbell shapes.

2.4 Measurement of Tensile Properties

The tensile properties were measured using an Instron machine Model: 3366, at a cross-head speed of 5 mm/min at $25 \pm 3^\circ\text{C}$ according to ASTM D 638-08. Tensile strength, tensile modulus, and elongation at break of every test samples were obtained from the average of five replicates with corresponding standard deviations.

2.5 Scanning Electron Microscopy (SEM)

The 1mm dumbbell fracture surface at failure mode from the previous tensile test was examined using Field Emission Scanning Electron Microscope (FESEM ZEISS SUPRA36VP-24-58). SEM micrographs were captured at various magnifications for morphological analysis. Before SEM process, a thin layer of platinum was used to sputter coat the fractured ends of the specimens which were mounted on aluminum stubs in order to avoid electrical charging during examinations.

2.6 Water Absorption

The water absorption test was conducted by immersing the samples in distilled water under ambient condition. 5 replicates which the size is kept nearly the same of every test samples were prepared. The test pieces of dimension 30mm x 5mm x 1mm were weighed by a Sartorius balance

Model: BS224S, with a precision of 1mg. A weight gains after exposure were recorded also every 4 hours for 3 days and followed by each day for another 57 days. The moisture content at any time t , M_t (%) as a result of moisture absorption calculated by using Equation 1.

$$M_t = 100 \times (w_t - w_o) / w_o \quad (1)$$

where w_o and w_t are original dry weight and weight after exposure, respectively.

2.7 Fourier Transform Infrared (FTIR)

The FTIR Perkin Elmer 2000 series instrument was used to determine the component and quality of sample by obtaining the FTIR spectra, according to ASTM E1252. The spectrum resolution was 4 cm^{-1} and the scanning range was from 650 to 4000 cm^{-1} .

3. Results and Discussion

3.1 Tensile Properties

The effect of fiber loading on the tensile strength of the untreated, NaOH treated and Silane treated PP/NBRr/BSP composites is shown in Figure 1. The results indicate that the addition of banana skin powders from 5 to 30 w/w decreases the tensile strength of all the treated and untreated PP/NBRr/BSP composites accordingly. Based on Figure 1, the untreated PP/NBRr/BSP composites have the lowest tensile strength, followed by NaOH treated composites and finally, α -APS silane treated composites. This may due to the incompatible of hydrophobic matrices (PP and NBRr) and hydrophilic nature of BSP [13] in the composite. Thus, the interactions of interfacial between fiber and matrices are weakened. The physical bonding (hydrogen bonding) formed inside the PP/NBRr/BSP composites after NaOH treatment and chemical bonding formed in the silane treated PP/NBRr/BSP composites improve interfacial adhesion between the PP/NBRr matrices and BSP fiber and thus results in higher tensile strength than untreated composite. Silane treated PP/NBRr/BSP composites exhibit highest tensile strength when compared to the PP/NBRr/BSP-NaOH and untreated PP/NBRr/BSP composites. This happens because the presence of coupling agent in silane treated PP/NBRr/BSP composites [9] has better stress distribution compared to the physical bonding formed in PP/NBRr/BSP-NaOH.

Figure 2 shows the effect of fiber loading on Young's modulus of the untreated, NaOH treated and Silane treated PP/NBRr/BSP composites. The results show Young's modulus of all treated and untreated composites increasing accordingly when the filler loading increased. The stiffness of the composites increased during the incorporation of BSP filler into the matrix. The addition of fiber as filler into the PP/NBRr matrices reduces the PP chains mobility and resulted increasing more rigid composites [9]. Meanwhile, Young's modulus for α -APS treated BSP composites has the highest values with increasing fiber loading. As explained earlier, better interactions between the matrix and filler achieved with the addition of silane coupling agent or undergoing NaOH treatment. A similar finding of previous research had also shown that higher Young's modulus in the surface of both NaOH treated jute fibers and modified jute fibers with silane coupling agent were caused by the higher rigidity of biocomposites [17].

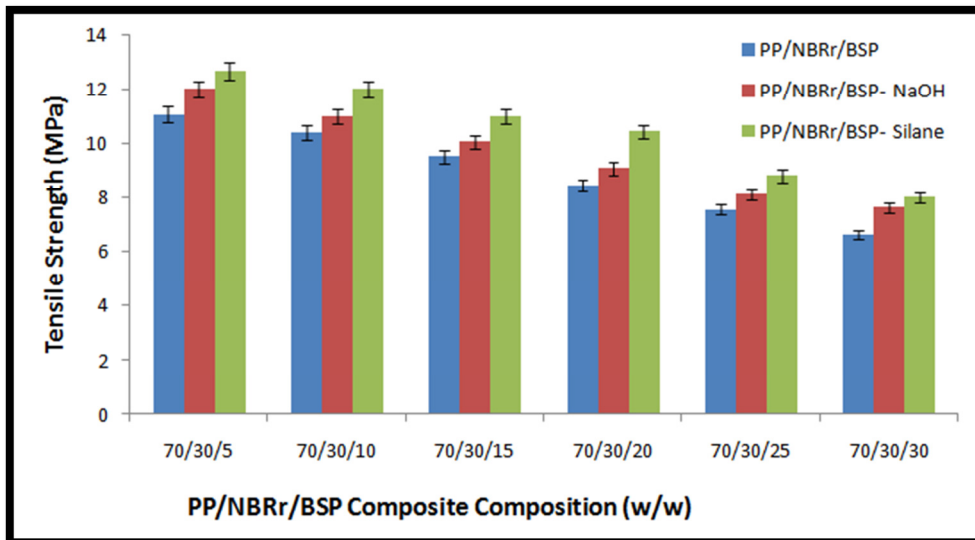


Fig. 1. Tensile Strength against fiber loading of PP/NBRr/BSP composites

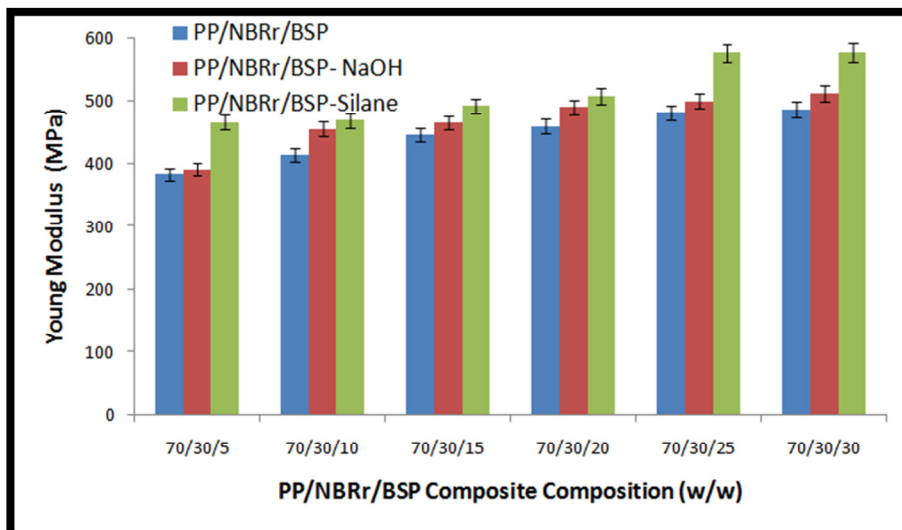


Fig. 2. Young Modulus against fiber loading of PP/NBRr/BSP composites

Figure 3 shows the elongation at break of the untreated, NaOH treated and Silane treated PP/NBRr/BSP composites. The results had shown that elongation at break of all composites decreasing with the increase of fiber loading. This is due to the increment of filler loading had hardened the composites and thus reduced their ductility. The values of elongation at break for PP/NBRr/BSP composites with α -APS treated BSP were the lowest, followed by composite with NaOH treated BSP and the highest values of elongation at break were the untreated composites. The elongation at break of α -APS treated PP/NBRr/BSP composites is lower than NaOH treated PP/NBRr/BSP composites because the physical bonding formed in NaOH treated PP/NBRr/BSP composites is weaker than the chemical bonding of α -APS treated PP/NBRr/BSP composites. Untreated composites have poor adhesion between polar banana skin powder and non-polar matrices resulted in lower elongation at break. Similar finding found which silane treated natural fiber results a stronger interfacial adhesion, consequences in low elongation at break in composites.

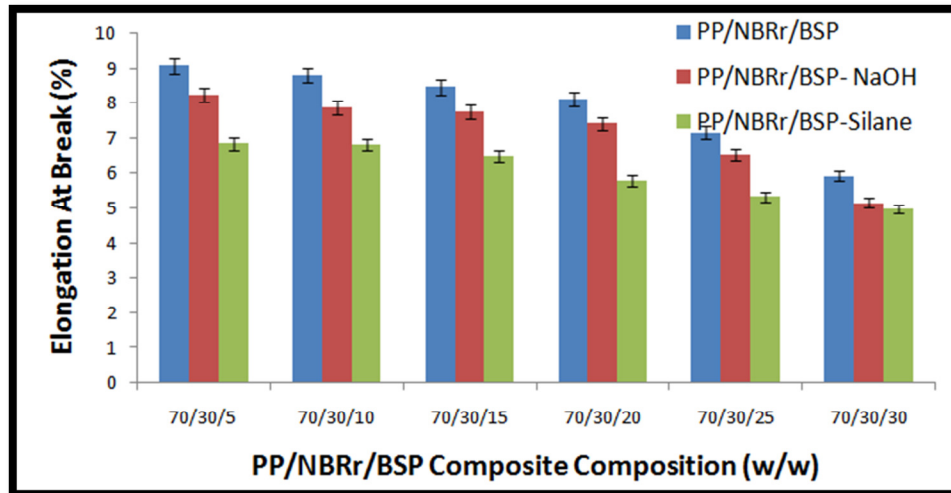


Fig. 3. Elongation at the break against fiber loading of PP/NBRr/BSP composites

3.2 Morphological Properties

Figure 4 (a–b) shows SEM micrographs of the fractured surface of untreated PP/NBRr/BSP composites (X500) with a filler loading of 5 and 30w/w. Figure 4 (a) indicates clear hole between the filler and matrix, which is the proof of poor adhesion between the filler and matrix. The lower adhesion can be seen in Figure 4 (b) which is the 30 w/w filler content composite as the BSP fibers were noticeably pulled out from the matrix and exhibited more detachments of BSP filler from the PP/NBRr matrices. Lower adhesion between the phases gives rise to poor stress transfer across the interface [5]. This proof that the higher the filler loading, the lower the strength of the composite.

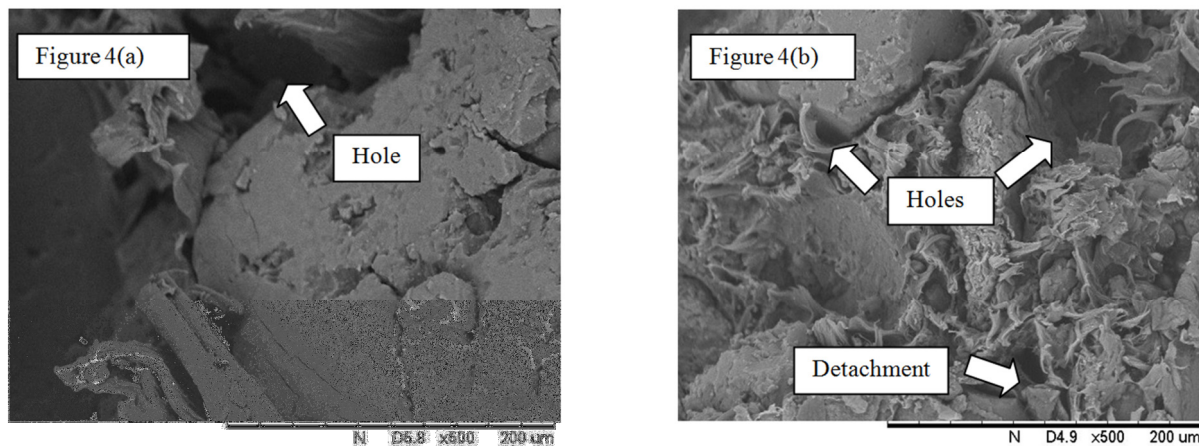


Fig. 4 (a-b). Scanning electron micrograph of the tensile fractured surface of untreated PP/NBRr/BSP at a magnification of 500x (a) 70/30/5 (b) 70/30/30

Figure 5(a–b) shows SEM micrographs of the fractured surface of NaOH treated PP/NBRr/BSP composites (X500) with a filler loading of 5 and 30w/w. By comparing Figure 5 (a-b), all the holes were filled with banana fibers for 5w/w filler content, while there were more cavities when increasing the filler content. This explained that the higher the fiber content, the weaker in strength for the composite. The removal of lignin by alkaline treatment was breakdown the fibre and led to

gaining of the effective surface area and surface charge exposed further hydroxyl and carboxyl groups [18]. Thus, the NaOH treated composites had better adhesion than untreated composites.

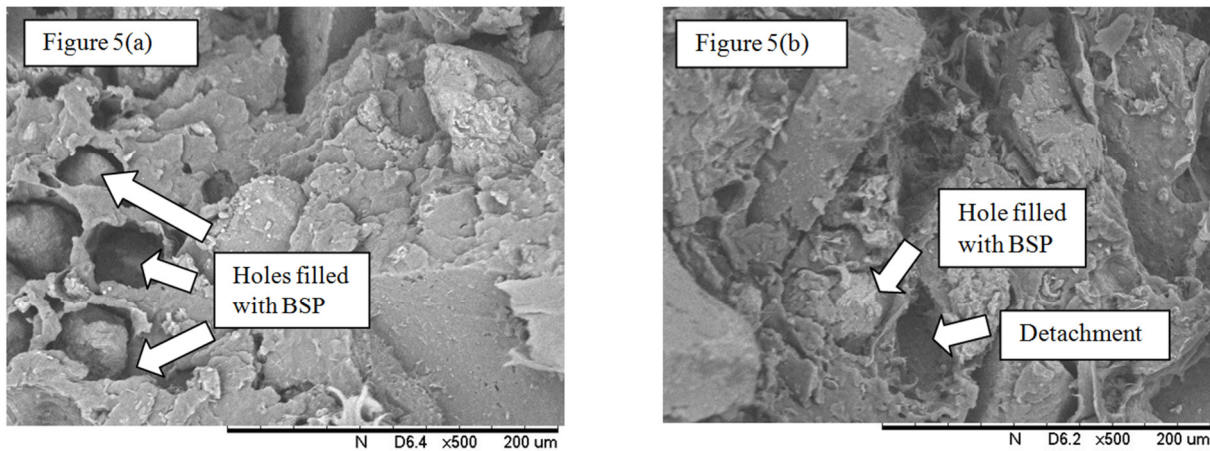


Fig. 5 (a-b). Scanning electron micrograph of the tensile fractured surface of PP/ NBRr/BSP-NaOH at a magnification of 500x (a) 70/30/5 (b) 70/30/30

Figure 6 (a–b) shows SEM micrographs of the fractured surface of α -APS treated PP/NBRr/BSP composites (X500) with a filler loading of 5 and 30w/w. It can be seen that α -APS treated PP/NBRr/BSP composites shows better attachment and dispersion of BSP filler and PP/NBRr matrices. The addition of a suitable coupling agent improved interfacial adhesion between hydrophilic fiber and hydrophobic matrices of the composite [9]. These statements are supported by the ductile morphology of the treated composites (Figure 6 (a–b)). The addition of coupling agent improves BSP fiber dispersion in the continuous PP and NBRr phase. The shear yield and tearing that occurred at the break of the composites can also be seen in Figure 6 as well. From Figure 6 (a-b), it can be observed that the higher the filler contents, the more detachments of BSP fiber occurred, the weaker in the strength of the composite.

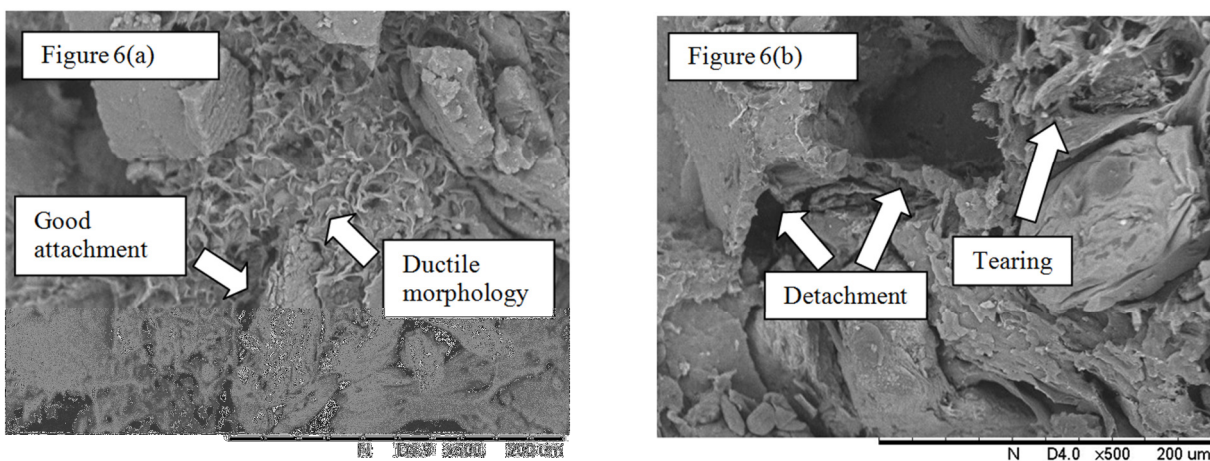


Fig. 6 (a-b). Scanning electron micrograph of the tensile fractured surface of PP/NBRr/BSP- α -APS at a magnification of 500x (a) 70/30/5 (b) 70/30/30

3.3 Water Uptake

Figure 7 shows the plot of PP/NBRr/BSP biocomposites with α -APS treated, NaOH treated and untreated BSP Water Uptake. The results show that the higher the BSP filler content in the

composite, the higher the water were absorbed by the composites. This is due to the formation of hydrogen bonds between water and BSP fiber as natural fibers consist of cellulose that contains polysaccharides which had hydroxyl group that is highly hydrophilic [9]. The number of hydrogen bonds between water molecules and organic components increased with filler content. This statement is supported by other research [19]. Based on Figure 7, the composites with NaOH and α -APS treatment had shown lower water absorption when compared to the untreated composites with similar filler content. This result shows that the BSP treatments with NaOH and α -APS separately have better interfacial interaction between BSP filler and PP/NBRr matrices than untreated composites, thus reducing the amount of equilibrium water absorption by the treated composites. This is due to α -APS silane as a coupling agent between fiber and matrices has the ability of the chemical to form a protective layer at the interfacial zone which results in preventing the direct diffusion of water molecules into the treated composites [4].

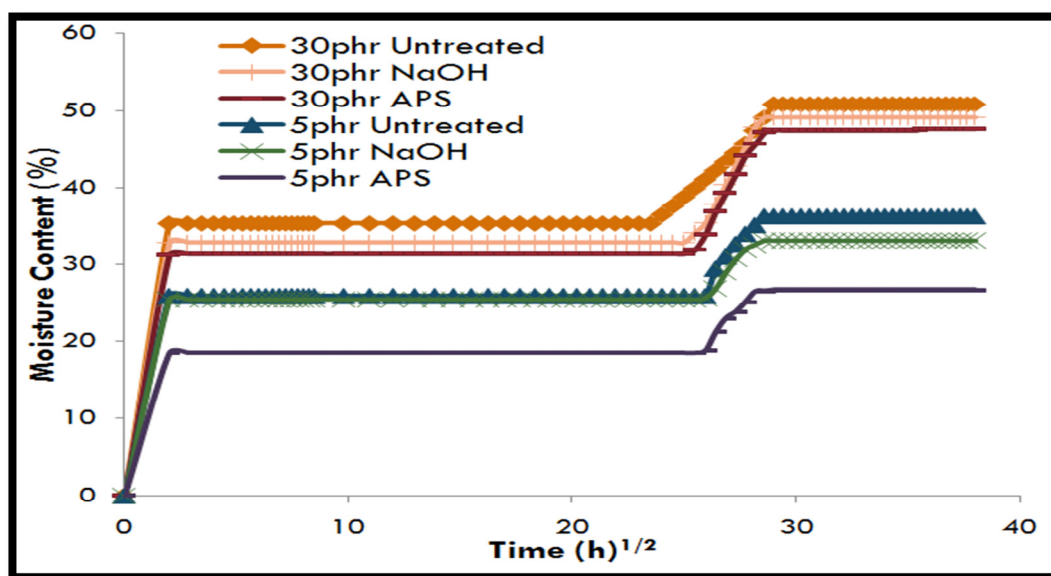


Fig. 7. Plot of PP/NBRr/BSP biocomposites with α -APS treated, NaOH treated and untreated BSP Water Uptake

3.4 FT-IR Spectroscopic Analysis

Figure 8 shows the FTIR spectra of BSP, NBRr and PP/NBRr/BSP composites (NaOH treated, α -APS treated and untreated). The spectra of PP/NBRr/BSP, NaOH treated PP/NBRr/BSP and α -APS treated PP/NBRr/BSP composites shows the OH stretching around $3700-3300\text{ cm}^{-1}$. The OH groups maybe were contributed by BSP filler. The OH groups have the deepest peaks for NaOH treated PP/NBRr/BSP composite. This is due to NaOH treatment causes the increment of the surface of the fiber to PP/NBRr matrices. The chemical mechanisms are shown in Figure 9. The peaks of stretching vibration of $\text{C}\equiv\text{N}$ group of NBRr appeared around $2300-2200\text{ cm}^{-1}$. $\text{C}=\text{O}$ group at ranged $1750-1700\text{ cm}^{-1}$ represents the ketone of hemicelluloses from BSP and can be seen removed after NaOH and Silane treatment at Figure 9. According to Figure 9, the stretching of the $-\text{Si}-\text{O}-\text{cellulose}$ is believed to occur at 1167.03 cm^{-1} which shows the presence of silane treatment in the composite. The chemical reactions of α -APS treatment on PP/NBRr/ BSP composites are shown in Figure 10.

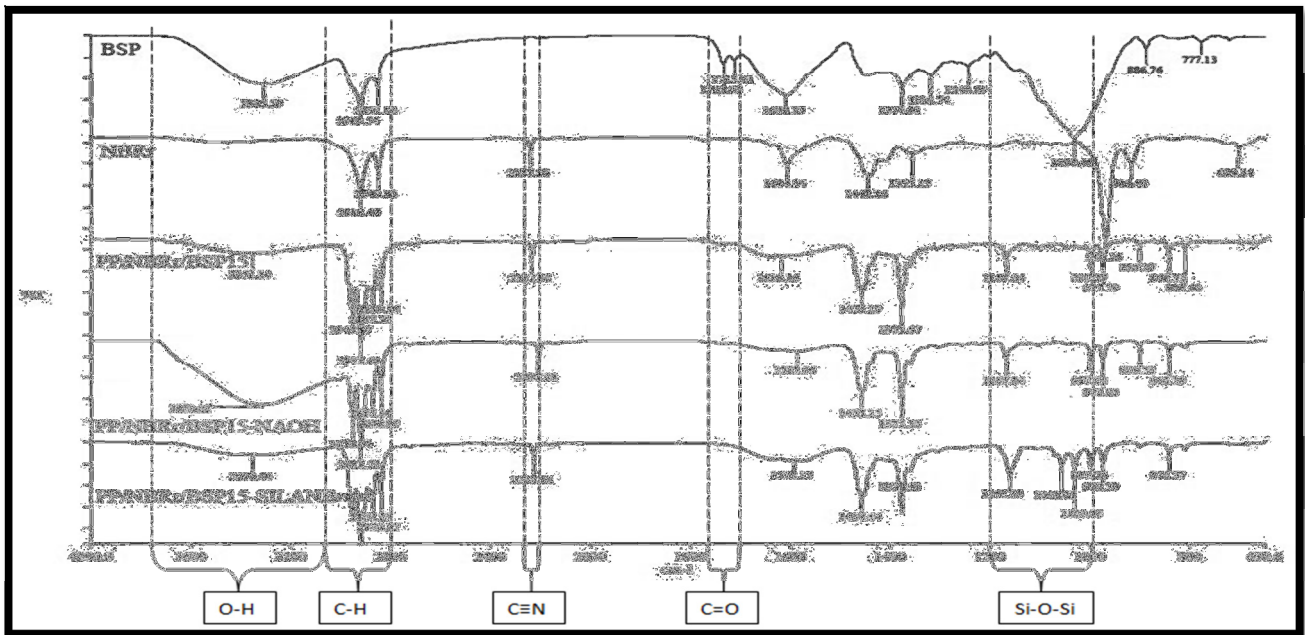


Fig. 8. FTIR spectra of BSP, NBRr and PP/NBRr/BSP composites (NaOH treated, α -APS treated and untreated)

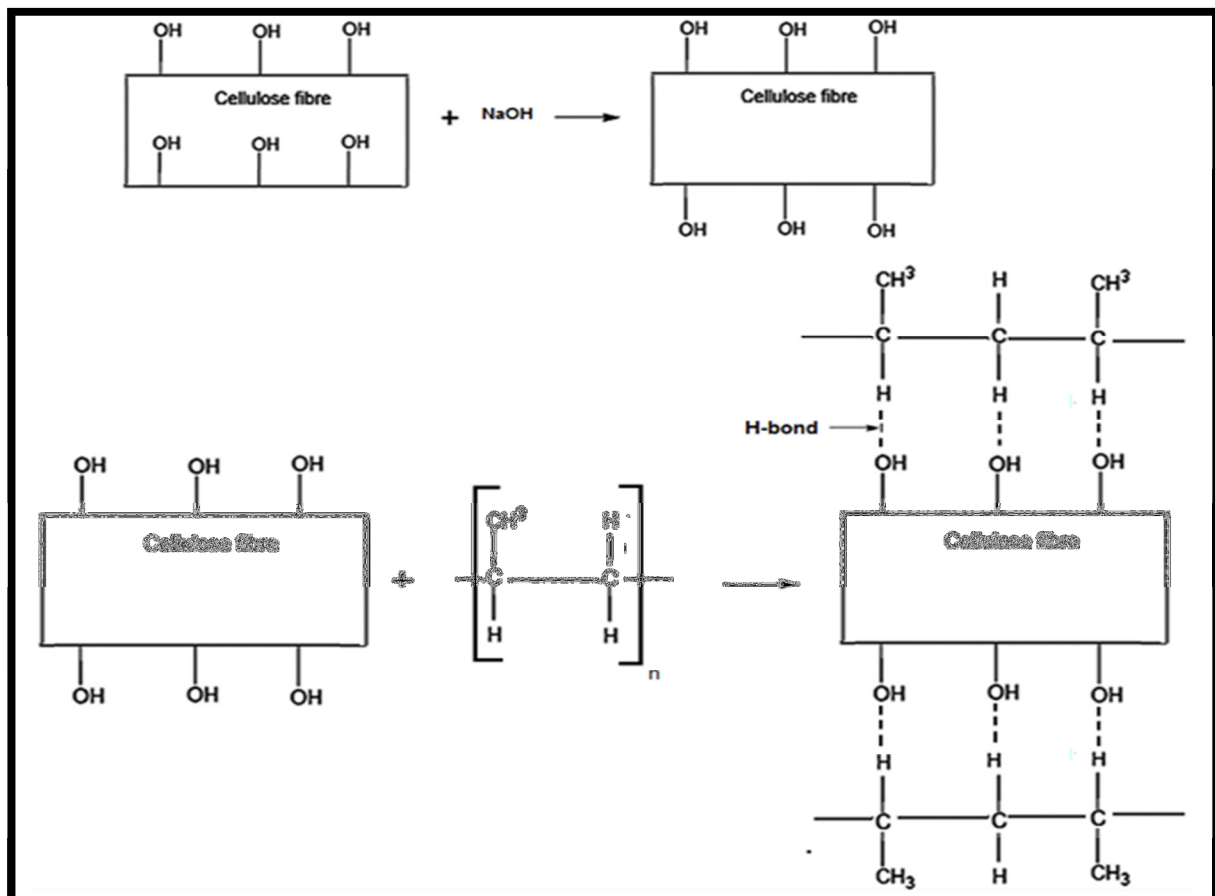


Fig. 9. General bond mechanism of NaOH treatment on the surface of BSP fiber

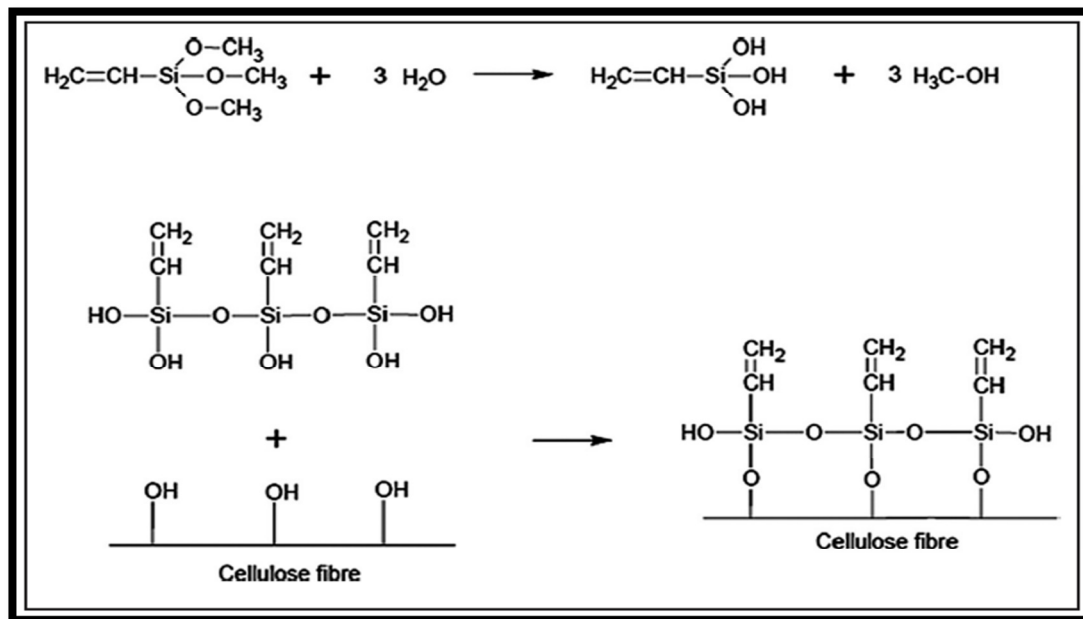


Fig. 10. General bond mechanism of silane coupling agent to the surface of fiber [20]

4. Conclusion

In conclusion, α -APS silane treatment have better improvement of mechanical properties than NaOH treatment and both treated PP/NBRr/BSP composites are better than untreated composites. The scanning electron microscopy shows that α -APS silane and NaOH treatment improved adhesion of BSP filler with PP/NBRr matrices. On the other hand, both α -APS silane and NaOH treated PP/NBRr/BSP composites have the same improvement of thermal properties and better than untreated PP/NBRr/BSP composites. DSC analysis shows α -APS silane treated composite has higher crystallinity portion than untreated composite might prove α -APS silane treated composite has better tensile properties as well. The FTIR spectrum of α -APS silane treated composite shows the presence of $-\text{Si}-\text{O}-\text{cellulose}$ that proven the bonding reaction in the BSP fiber.

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