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Influence of 3-aminopropyltrimethoxysilane on the Mechanical and Morphological Properties of Polypropylene/ Acrylonitrile Butadiene Rubber Blend Filled Kenaf Fibre

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ABSTRACT

In this paper, the effect of 3-aminopropyltrimethoxysilane (APS) as coupling agent on the mechanical and morphological properties of polypropylene (PP)/ acrylonitrile butadiene rubber (NBR)/ kenaf fibre (KF) composites were investigated. Kenaf fibre was treated with 5 % of APS with different fibre loading (5, 10, 15, 20 and 30 phr). The mixture of PP/NBR/kenaf was done using a heated two-roll mill at 180 °C for 9 minutes followed by compression moulding by hot press machine to produce 1 mm thin sheet. With the dumbbell shape specimens, tensile test and morphological study was done using Instron Universal Testing Machine (UTM) and Scanning Electron Microscope (SEM), respectively. From the data obtained, the tensile strength was decreased as kenaf fibre loading increased. However, for the composites with silane treatment showed increment in tensile strength. This was due to the better interfacial adhesion between fibre and the matrix. This also has been proved by morphological study as good attachment and better dispersion of fibre was observed for the treatment composites.

1. Introduction

Thermoplastic elastomer (TPE) is the combination of thermoplastic polymer and rubber that has attracted much attention in polymer field. TPE have excellent properties such as simple compounding and easily recyclable [1,2]. Polypropylene (PP) is a polyolefin type made up from propylene monomer with linear hydrocarbon. It is widely used in packaging, containers, laboratory equipment and others. PP can be combined with other materials such as rubber, to make it more pliable. Acrylonitrile butadiene rubber (NBR) has been widely used in oil seals and automotive host due to its low cost.

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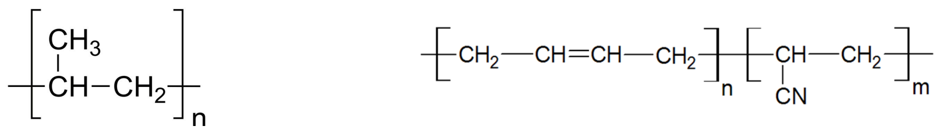
NBR also has excellent resistance to oil, fuels and greases, and easy to process. The properties of NBR is depends on the acrylonitrile content. Figure 1 show the chemical structure of PP and NBR.



(a) (b)

Fig.1. Chemical structure of (a) polypropylene and (b) acrylonitrile butadiene rubber [3]

The serious concern that affects to the environment is abundance of plastics produced. Most of the plastics are not biodegrade and difficult to decompose. Thus, the current percentage of plastics disposal at the landfill kept increasing. This has attracted the scientists, researchers and industries to address this matter by using a natural fibre replacing synthetic fibre in order to decrease the use of petroleum based materials and to produce green products. Many studies have been done on the potential of natural fibre in various polymer composites [4,5]. Natural fibre composites have their own advantages such as high availability, renewable and low in cost. In this study, kenaf fibre was selected as filler for polymer and rubber composites.

The addition kenaf fibre has taken into consideration. This is because kenaf as a natural fibre is not compatible with hydrophobic polymer matrix. The poor compatibility of hydrophobic polypropylene and hydrophilic kenaf leads to the formation of weak interfaces between them. This will obstruct from getting a good physical and chemical interaction across the phase boundaries. Therefore, chemical treatment or surface modification of natural fibre is required to improve the adhesion between matrix and filler and able to increase the stress transfer from the polymer matrix to the filler.

The objective of this study is to evaluate the mechanical and morphological properties of PP/NBR/kenaf with silane treatment. 3-aminopropyltrimethoxysilane (APS) was used as the chemical treatment for kenaf fibre.

2. Material and Methods

2.1 Materials Preparation

Isotactic polypropylene (PP) used in this work was supplied by Titan PP Polymers Sdn Bhd (code 6331) with density 0.9g/cm^3. The acrylonitrile butadiene rubber (NBR) was obtained from Zarm Scientific & Supplies Sdn Bhd, Bukit Mertajam, Penang. Kenaf fibre was supplied from Lembaga Kenaf & Tembakau, Kelantan. 3-aminopropyltrimethoxysilane (APS) was supplied by Sigma Aldrich Chemical. Firstly, kenaf was dried in the oven with temperature 85°C for 24 hours to remove moisture content. Then, kenaf was grinded and sieved into powder form with particle size less than 300 µm. Kenaf was dried again to remove excessive moisture.

2.1.1 APS silane treatment

The chemical modification of kenaf fibre was carried out at room temperature. Kenaf fibre was immersed in 5% alkaline solution for 2 hours and washed with tap water for a several times to remove excess solution. 1% of diluted acetic acid was discharged to remove any traces from the surface of fibre. Alkaline-treated kenaf was washed again with distilled water and dried at room temperature for 3 hours prior put it in the oven for 24 hour. Then, 5% of 3-aminopropyletrimethoxysilane was prepared in acetone solution with addition of acetic acid to maintain the pH solution by 4. Silane-treated kenaf was remove from the solution after soaked for 1 hour and dried again in the oven for 48 hour. The samples was crushed and sieved to gain homogeneous particle size of fibre.

2.2 Preparation of Composite

Table 1 show the formulation used for PP/NBR/KF composites. A heated two-roll mill machine (Model DW-5110) from Fang Yuan Instrument (DG) Co. Ltd, China, was used to melt-mixed the materials. The machine was set up at temperature 180°C. The speed of the machine was adjusted to 15 rpm. PP was loaded first and melts for four minutes, followed by NBR and mixed for another two minutes. Kenaf was added at minute sixth and the compound was mixed until minutes ninth. During the melting period, scrapping process need to be continuously proceeded to ensure the compound was mixed very well. The mixtures were compressed using a hot press, model GT-7014 at 180°C for 11 minutes. Preheating was allowed for seven minutes, two minutes for compression and another two minutes for cooling pressed. 1mm thin sheet was cut into the dumbbell shape using a Wallace die cutter S6/1/6.A.

Table 1
Formulation of PP/NBR/KF composites

Composite materials	Materials Ratio (phr)					
PP	70	70	70	70	70	70
NBR	30	30	30	30	30	30
Kenaf or Silane-Treated Kenaf	0	5	10	15	20	30

2.3 Composite Characterization
2.3.1 Tensile test

Tensile test was one of the mechanical properties evaluated in this study. It was conducted using ASTM D638 using Instron tensile machine model no 3366. Dumbbell shape specimens of 1 mm thickness were utilized for tensile testing. Tensile strength, elongation at break and Young's modulus were measured at a crosshead speed of 5 mm/min. Five specimens were used for each test and the average data with a corresponding standard deviation was reported.

2.3.2 Morphological Study

The scanning electron micrograph was obtained using a JOEL JSM-6460LA. Prior to SEM observations the kenaf raw materials and the fractured ends of the specimens were sputter coated with a thin layer of platinum to avoid electrical charging during examinations.

3. Results and Discussion
3.1 Effects of Kenaf Fibre Loading and Silane Coupling Agent on Mechanical Properties

Figure 2 show the effects of kenaf fibre loading and silane coupling agent on tensile strength of PP/NBR/KF composites. As the kenaf loading increase, the tensile strength slightly decrease. The reduction may due to the poor dispersion of kenaf fibres. According to Ismail *et al.*, [6], the incorporation of natural plant fibres into polymer matrix might increase or decrease the composites strength. These was mainly depends on the shape of the fillers used. For the irregularly shaped fibres, the composites strength mostly decreases due to the inability of the fibres to support the stressed transferred from the polymer matrix. Figure 5 (a) show the SEM micrograph of kenaf fibre applied to this work. As can be seen, kenaf fibres represent multiple shapes and sizes that liable for the decline of tensile strength. However, with silane coupling agents, tensile strength resulted to be improved by 5% to 20% with different fibre loading. The improvement was due to the strong affinity of amino groups of silane coupling agent towards the hydroxyl groups of natural fibres. Xie *et al.*, [7] has reported that the natural fibre surface turns to be more hydrophobic and able to bind with the polymer matrix [7].

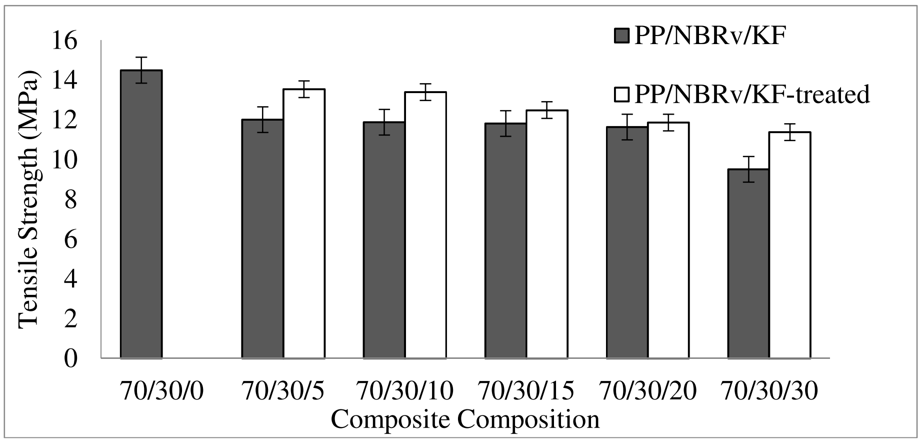


Fig. 2. Tensile strength of composite with and without silane treatment

Figure 3 show the effects of kenaf fibre loading and silane coupling agent on Young's modulus of PP/NBR/KF composites. The Young's modulus slightly increases as increasing the kenaf loading. Previous study from other researchers also reported that the modulus value is increases as the fibre loading increase [8]. As more fibre was added, the stiffness of composites become higher depends on the fibre properties. For silane-treated kenaf composites, the modulus is significantly increase due to the better adhesion formed between both of the fibre and matrix.

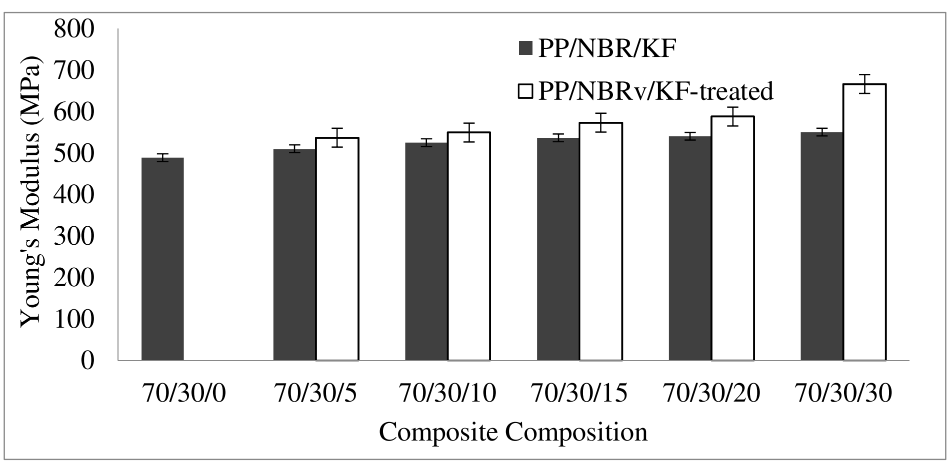


Fig. 3. Young's modulus of composite with and without silane treatment

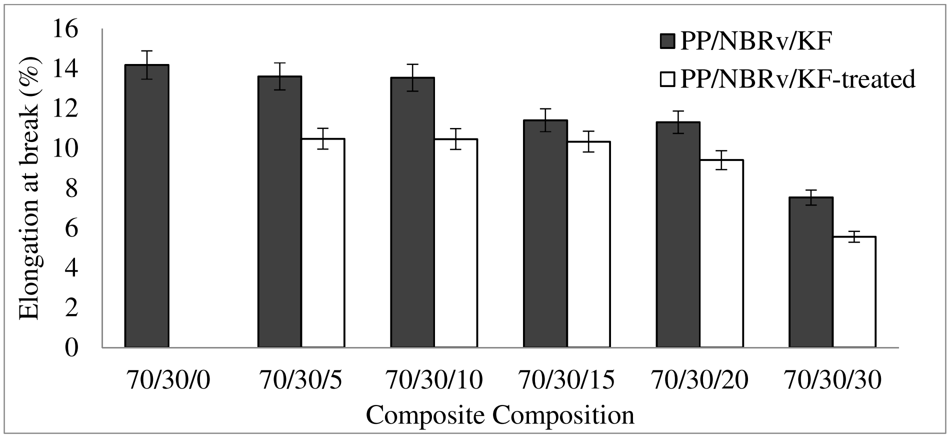


Fig. 4. Elongation at break of composite with and without silane treatment

Figure 4 show the effect of kenaf fibre loading and silane coupling agent on elongation at break, E_b of PP/NBR/KF composites. As expected, E_b of PP/NBR/KF composites decreases with increasing fibre loading. According to Razavi-nouri *et al.*, [9], at higher filler loading, there is a restriction of the polymer molecules from mobilized to each other. However, for the silane-treated kenaf composites, the E_b is much lower. The presence of silane coupling agent have improved the surface functionality of kenaf fibres and made it effectively bonded with the polymer matrix.

3.1 Effects of Kenaf Fibre Loading and a Silane Coupling Agent on Morphological Properties

Figure 5 (a) show the micrograph of kenaf fibre raw which has long fibres and smooth fibre surface. Prior to silane treatment, kenaf fibre was immersed in an alkaline solution. Via alkaline treatment, the fibre surfaces become more roughness due to the cellulose exposes on the fibre surface which increase the potential of chemical bonding [10]. With the silane treatment, the silanol reacts with OH groups of the cellulose of kenaf fibres which can increase the degree of crosslinking by increasing the surface area. Figure 5 (b) show the silane-treated kenaf. It can be seen from the enlarged micrograph, the kenaf fibre bundle showed opened fibres or also known as elementary fibres and expected to form fibrillation. This will increase the fibre surface area and allowing for stronger bonding between fibres and matrix.

Figure 5 (c-f) shows the micrograph of tensile fracture surface of kenaf fibres filled PP/NBR composites with and without treatment at 5 and 30 phr, respectively. It can be seen from Figure 5 (c and e), PP/NBR/KF composites without treatment showed some holes due to the pulled out fibre from the matrix. There are also presented non-homogeneous of kenaf fibre as well as unwetted fibre on the polymer matrix. The poor adhesion and weak bonding of kenaf fibre make it detach from the PP/NBR matrix. However, for PP/NBR/KF-treated composites shows from Figure 5 (d and f), it can be observed, less detachment site and less gap presented between the fibres and matrix. A better adhesion and improved wetted between the kenaf fibres and PP/NBR matrix were observed (showed by the arrows). This has been enhanced the tensile properties of composites as discussed earlier.

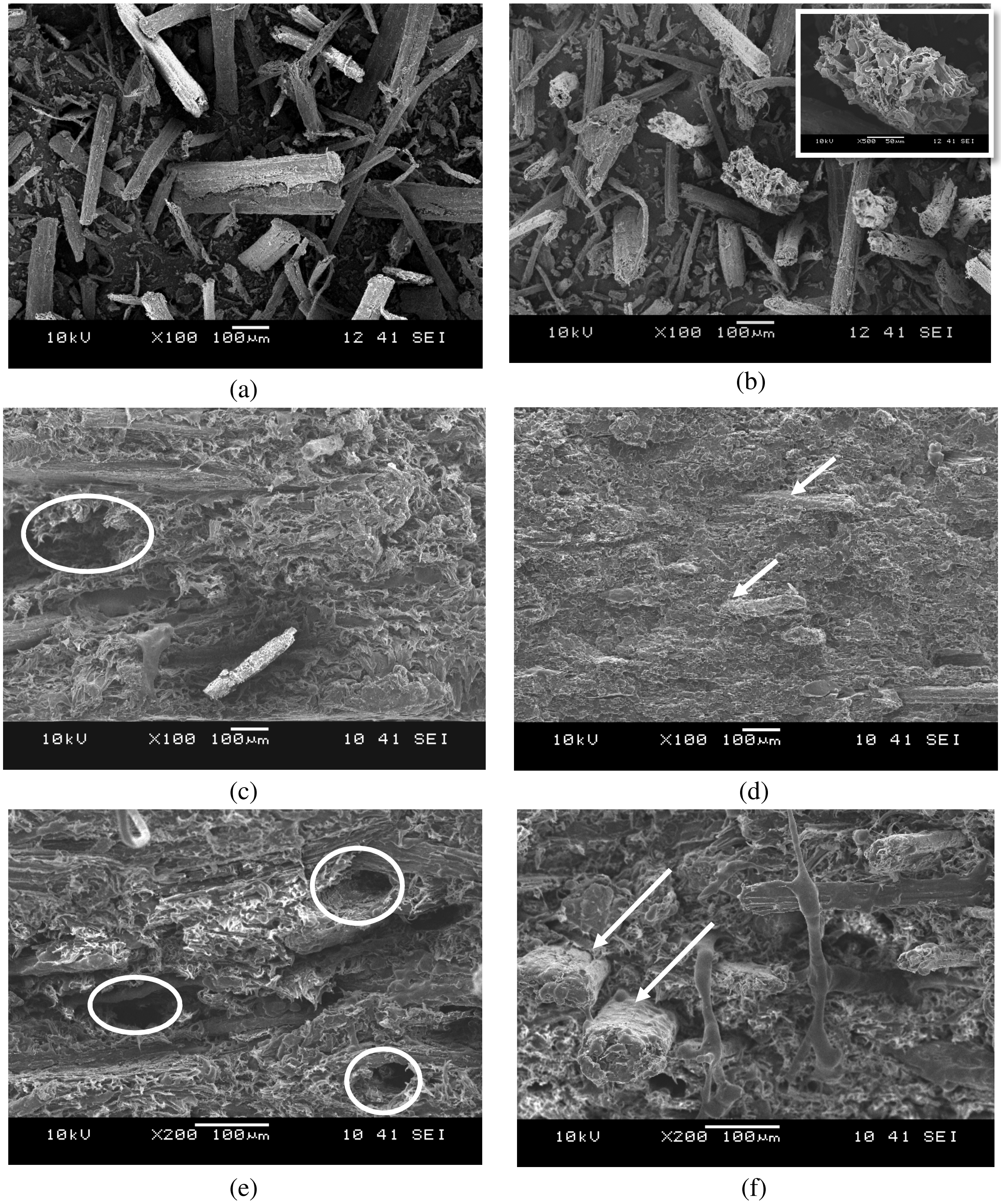


Fig. 5. SEM micrograph of (a) kenaf raw; (b) silane-treated kenaf; (c) PP/NBR/kenaf composite with 5 phr (X 100 magnification); (d) PP/NBR/kenaf-treated with 5phr (at X100 magnification; (e) PP/NBR/kenaf with 30 phr (X200 magnification); (f) PP/NBR/kenaf-treated with 30 phr (X200 magnification)

4. Conclusion

The tensile strength and elongation at break decrease with increasing the kenaf fibre loading but decreasing the Young's modulus. The presents of silane as coupling agent improved the tensile strength as well as the modulus. Better adhesion resulted to allow of stress transfer from the PP/NBR matrices to the kenaf fibre. SEM micrograph proved an improved of adhesion between the kenaf fibre and PP/NBR matrix with silane treatment.

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