



## THE COMPATIBILIZING EFFECT OF POLYPROPYLENE MALEIC ANHYDRIDE (PPMAH) ON POLYPROPYLENE (PP)/ACRYLONITRILE BUTADIENE RUBBER (NBR)/PALM KERNEL SHELL (PKS) COMPOSITES

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### ABSTRACT

The compatibilizing effect of polypropylene maleic anhydride (PPMAH) on the mechanical and morphological properties of polypropylene (PP)/ acrylonitrile butadiene rubber (NBR)/palm kernel shell (PKS) composites were studied. PP and NBR are found highly incompatible and can be improved by compatibilizer such as PPMAH. The composites were melt mixed using heated two roll mill at 180°C and speed of 15 rpm with six different loading (100/0/10, 80/20/10, 70/30/10, 60/40/10, 50/50/10, 40/60/10 phr) with fixed 5 phr PPMAH. Increasing of NBR loading in PP/NBR/PKS composites decreases the tensile strength and Young's modulus but increases the elongation at break. The results showed that higher tensile strength and Young's modulus were obtained for PPMAH compatibilized composites as compared to uncompatibilized composites. Tensile fractured surfaces observed by scanning electron microscopy (SEM) indicates improved adhesion of palm kernel shell with polypropylene / acrylonitrile butadiene rubber matrices in the presence of polypropylene maleic anhydride.

**Keywords:** polypropylene maleic anhydride, Compatibilizer, acrylonitrile butadiene rubber, palm kernel shell, composites.

### INTRODUCTION

In recent years, elastomeric rubber-plastic mixture has become technologically important for use as thermoplastic elastomer (TPE). The objective of the mixture of plastic and rubber is the improvement of physical, thermal and mechanical properties as well as the modification of processing characteristics and cost reduction of the final product (Ismail *et al.*, 2004).

Polypropylene (PP) is one of the commonly employed polyolefin because of its low price, stabilized properties, and simple processability. Acrylonitrile butadiene rubber (NBR) is well known unsaturated copolymers for about five decades. It has been used in many industrial required purposes as hoses, O-ring seals, insulation base product and other many packaging materials. It is a commonly used as oil resistant rubber (Ahmed, 2014). Currently, the natural fillers were added to elastomers to obtain cheaper and lighter composite materials.

Natural fillers have many benefits such as increased stiffness, thermal resistance, stability and strength (Gupta and Gupta, 2005). Natural fillers are potential candidate as solution of environmental burden from oil palm industry. Oil palm industry in Malaysia produced over 100 million tons of biomass such as palm kernel shell and palm pressed fiber with its 6 million hectares of plantation (Khalil *et al.*, 2012). This huge amount of palm kernel shell waste can be solved by using palm kernel shell as filler in composite materials.

The mixture of polypropylene (PP) / acrylonitrile butadiene rubber (NBR) combines the oil resistance of NBR as well as excellent chemical and moisture resistance, good ductility and stiffness, low density, and easy processing characteristics of PP. Actually, PP and NBR are highly incompatible and usually exhibit phase separated morphology because of poor physical, mechanical and chemical interactions across the phase boundaries (Xu *et al.*, 2013). Hence, this properties can be further improved with suitable compatibilization techniques (Lee *et al.*, 2009). Polypropylene maleic anhydride (PPMAH) can be used for compatibilizer, due to its ability to compatibilizing polypropylene polymer matrices (Seo *et al.*, 2000). George *et al.* (1995) reported that for polypropylene maleic anhydride compatibilized composites, the increase in tensile properties is due to the increased dipolar interaction between the PPMAH and NBR phases, which cause an increase in interface adhesion between PP and NBR phases. To our knowledge, no investigations have been carried out on the compatibilizing effect of PPMAH in the PP/NBR/PKS composites. So in this present work, compatibilization of PP/NBR/PKS composites, using polypropylene maleic anhydride (PPMAH) are reported and effects on the mechanical, morphological properties and chemical interaction were evaluated.

### EXPERIMENTAL

#### Materials



The materials used for the preparation of PP/NBR/PKS composites are shown in Table-1. Palm kernel shell (PKS) was dried under the hot sun for 24 hours and then put into an oven at 90 °C for 3 hours to remove moisture. A pulverized machine model RT-34 from Rong Tsong Precision Technology Co and sieved from Endocotts sieve were utilized to obtain an average particle size of 150-300 µm. NBR was then cut and resized into small pieces.

**Table-1.** Materials specification and description.

Material	Source
Polypropylene (PP)	Titan Pro Polymers (M) Sdn. Bhd.
Acrylonitrile Butadiene Rubber (NBR)	Zarm Scientific and Supplier Sdn. Bhd.
Palm Kernel Shell (PKS)	Solid Orient Holdings Sdn.Bhd, Kedah, Malaysia
Compatibilizer (polypropylene maleic anhydride)	Alfa Aesar (M) Sdn. Bhd.

### Formulations

The formulations of the PP/NBR/PKS composites components are as presented in Table-2. PP/NBR/PKS composites with and without PPMAH were evaluated.

**Table-2.** Formulations of PP/NBR/PKS composites components.

Materials	Loading (phr)					
PP	100	80	70	60	50	40
NBR	0	20	30	40	50	60
PKS <sup>a</sup>	10	10	10	10	10	10
PPMAH <sup>b</sup>	5	5	5	5	5	5

<sup>a</sup> phr of resin.

<sup>b</sup> phr of pp.

### Preparation of the composites

The composites with different loading prepared by using heated two roll mill machine from Fang Yuan Instrument (DG) Co. Ltd at 180 °C. The heated two roll mill was allowed to rotate slowly at 15 rpm for homogeneous heat distribution between the rollers. PP was charged into the machine and was allowed to be melted for 2 min prior to NBR addition. This mixture was allowed to be mixed for another 4 min. PKS was added at the 6<sup>th</sup> min and continued to be mixed homogeneously until the 9<sup>th</sup> min. This composite were removed and kept in desicator to remove moisture content. Similar mixing procedures were undertaken for PPMAH compatibilized composites.

### Compression moulding

1 mm thin sheet were form with the help of a hot pres model GT 7014 A 6895 kPa at 180°C. A 7 min preheating and 2 min compression followed by 2 min cooling was carried out. The thin sheet was further cut into a dumbbell shape using a wallace die cutter model: S6/1/6.A.

### Tensile properties

Tensile test was carried out by utilizing an universal tensile machine Instron 5569 at 25 °C according to ASTM D381. The cross head speed was fixed at 5 mm/min. Tensile strength, Young's modulus and elongation at break were measured. Five repetitive dumbbell specimens were employed for tensile properties evaluation.

### Morphological properties

Morphological studies on tensile fracture surfaces of all compositions were carried out using a Scanning Electron Microscopy (SEM), model JOEL JSM-6460LA. Tensile fracture surface specimen was mounted on aluminium stubs and was coated with gold to avoid electrostatic discharge during examination.

### Fourier Transform Infrared (FTIR) analysis

Fourier Transform Infrared (FTIR) analysis was used to evaluate the chemical composition of NBR, PKS, PP/NBR/PKS composites and PP/NBR/PKS-PPMAH compatibilized composites. Solid and sheet form samples



were cut into small rectangular sheet and used directly with the FTIR machine. FTIR spectra were recorded by plotting wavelength against absorbance. The spectrum resolution was  $4\text{ cm}^{-1}$  and the scanning range was from  $650$  to  $4000\text{ cm}^{-1}$ .

## RESULTS AND DISCUSSIONS

### Tensile properties

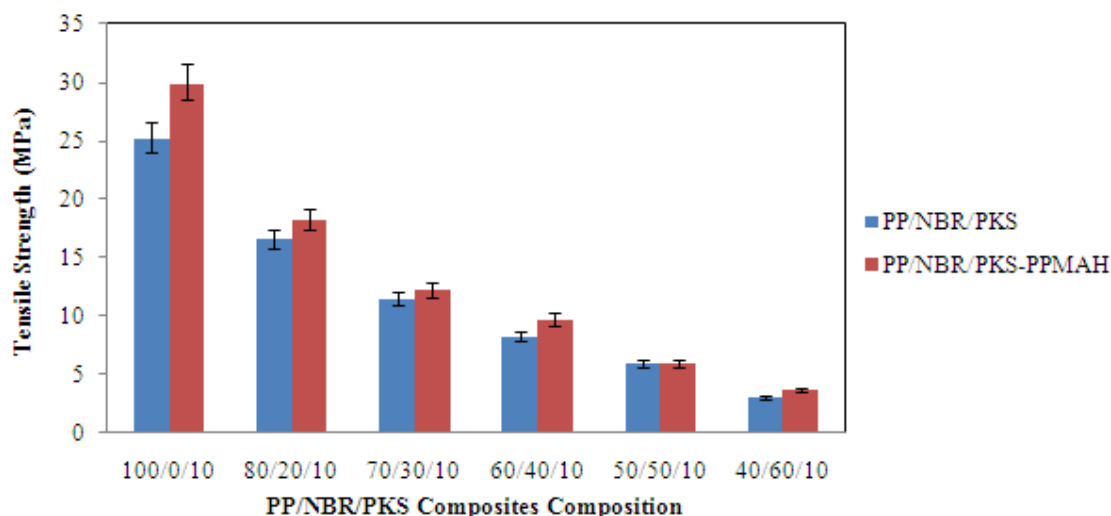
Figures 1 and 2 shows the tensile strength and Young's modulus of PP/NBR/PPMAH composites with and without PPMAH compatibilization. It has found the both properties decrease with increasing NBR loading. This is not surprising since the addition of NBR leads to decrease in rigidity of the resulting composites (Ismail and Suryadiansyah, 2002). And this may due to the presence of the soft rubber phase and fall in crystallinity of the PP phase (George *et al.*, 1995).

The tensile strength and Young's modulus of composites with PPMAH is found to be higher than the uncompatibilized composites. Incorporation of PPMAH

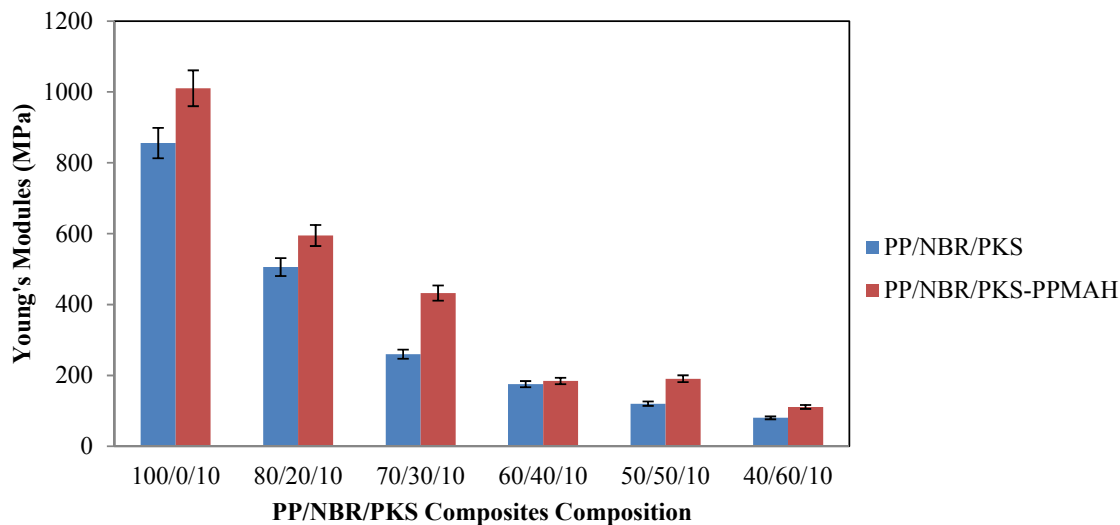
further improves the mechanical properties of the composites due to the further increased dipolar interaction between PPMAH and NBR phases (Xu *et al.*, 2013). Besides, anhydride groups reacts with hydroxyl groups of PKS powder and form ester bonds, improving the interfacial adhesion between the matrices and filler (Zhou *et al.*, 2013). Similar chemical interaction was reported by Salleh *et al.*, (2014) on maleic anhydride (MAH) modification with different filler.

However further addition of NBR in the composite lead to poor mechanical properties due to decrease of PP portion in the composite. Xu *et al.*, (2013) have reported that the tensile strength of composites decreased with increasing of NBR in the composite using glass fibers.

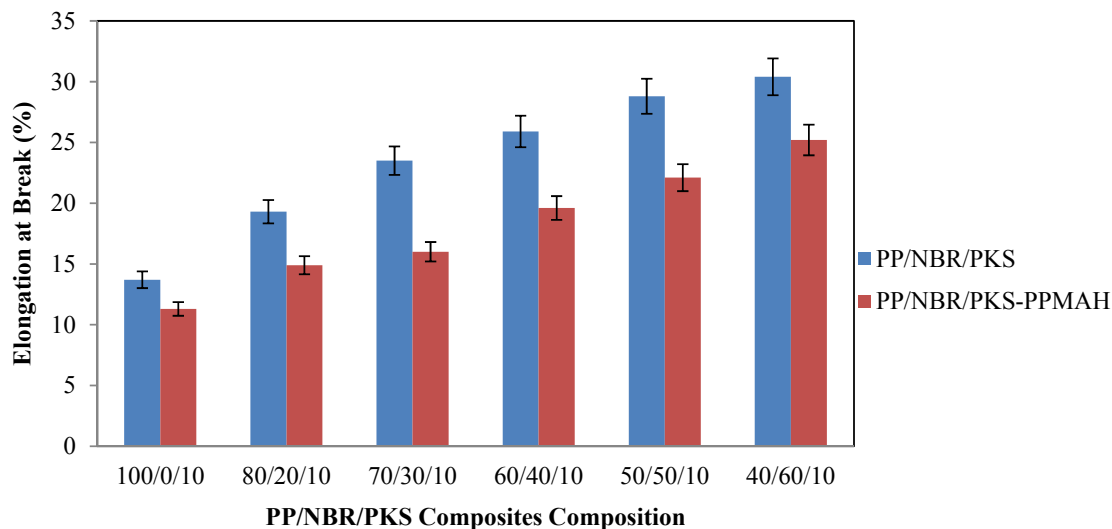
The tensile strength of a composite mainly depends on the crystallinity of PP and rigidity of the material in the presence of filler. With the increase of elastomer loading, the rigidity of PP continuous phase decreased, which resulted in the decreased strength of the composite.



**Figure-1.** Tensile Strength of PP/NBR/PPMAH composite with and without PPMAH compatibilization.



**Figure-2.** Young's Modulus of PP/NBR/PKS composites with and without PPMAH compatibilization.



**Figure-3.** Elongation at Break of PP/NBR/PKS composites with and without PPMAH compatibilization.

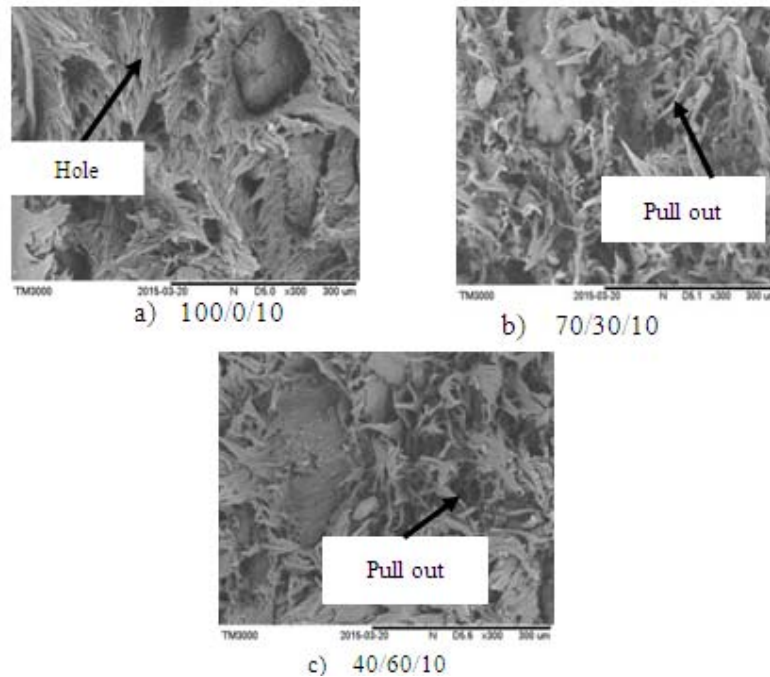
Figure-3 shows the elongation at break of PP/NBR/PKS composites with and without PPMAH compatibilization. The elongation at break increases with increasing of NBR loading in both composites. This may be due to addition of NBR reduces the stiffness of the composites which resulted in lower resistance to elongation (Ismail *et al.*, 2009).

#### Morphological properties

Figures 4 and 5 shows micrographs of tensile fracture of uncompatibilized and compatibilized PP/NBR/PKS composite at 300x magnification. Based on the SEM micrographs, the fracture surfaces has filler pull

out as a result of weak interface between PKS fillers and PP/NBR matrices. This may indicate poor adhesion and compatibility between PKS filler and PP/NBR matrices. However in PPMAH compatibilized composites SEM micrograph shows better dispersion of PKS particles in PP/NBR matrices.

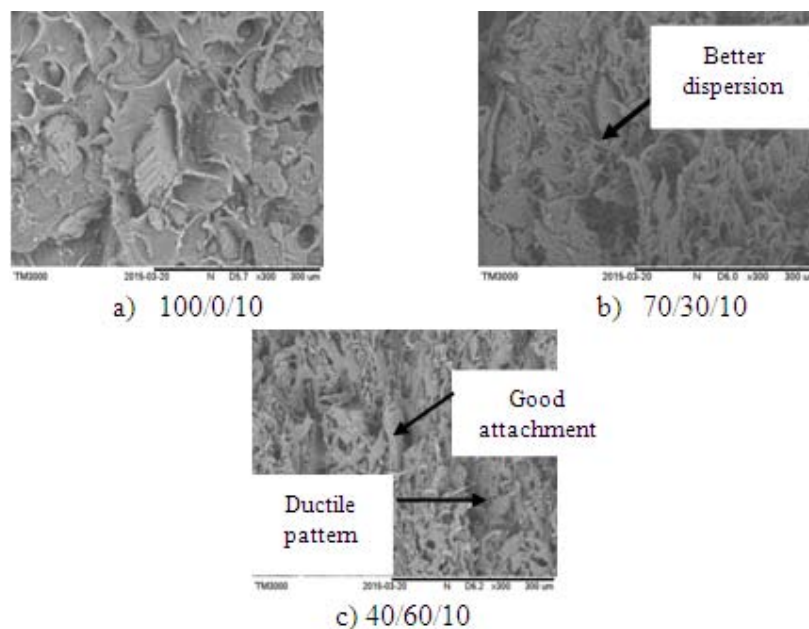
Figure-4(a) to (c) indicates presence of multiple holes and pull outs due to weak adhesion between the PP/NBR matrices and PKS filler. More pull out of PKS filler in uncompatibilized composite which may be due to unbounded PKS filler in the composites. Poor adhesion between the phases may result to rises to poor stress transfer across the interface.



**Figure-4.** Micrographs of tensile fracture of uncompatibilized PP/NBR/PKS composite at 300x magnification.

From Figure-5 it can be seen that the adhesion and better dispersion between PP/NBR matrices and PKS filler was enhanced in the presence of PPMAH. The PKS filler were well bonded to the composites in the presence

of PPMAH. It is believed to increase the attachment between filler and matrix. Simillar findings have been reported by Santiago *et al.* 2012 on PP/NBR matrices using different fillers.



**Figure-5.** Micrographs of tensile fracture of compatibilized PP/NBR/PKS composite at 300x magnification.

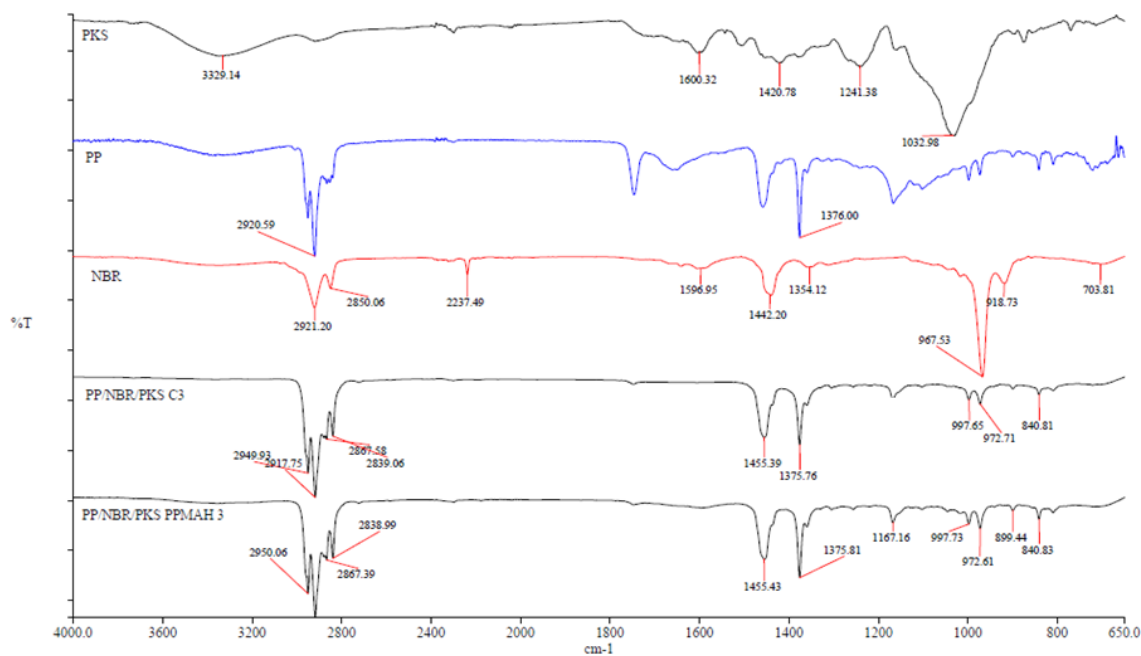


### Fourier Transform Infrared Spectroscopy (FTIR) analysis

Figure-6 shows FTIR spectra comparison of PKS, NBR, PP/NBR/PKS, and PP/NBR/PKS-PPMAH compatibilized composites in the region of 650 to 4000 $\text{cm}^{-1}$ . Nitrile stretch ( $\text{C}\equiv\text{N}$ ) was found around 2237  $\text{cm}^{-1}$  for Acrylonitrile Butadiene Rubber (NBR) because it is an important reactive site for cleavage of bonds to be occurred. However, the absence of nitrile stretch peak for composites of PP/NBR/PKS and PP/NBR/PKS-PPMAH

compatibilized composite could be due to the reaction happened between PP, NBR and PKS.

The spectra of both PP/NBR/PKS and PP/NBR/PKS-PPMAH composites show the OH stretching around 3300  $\text{cm}^{-1}$ . The OH groups may be contributed by PKS filler. In PPMAH compatibilized composite, the absorption of C-O-C around 1050-1250 $\text{cm}^{-1}$  indicated the presence of ester groups was observed. This is due to the formation of bonding between MAH moiety with PKS filler in the presence of PPMAH.



**Figure-6.** FTIR spectra for PKS, NBR, PP/NBR/PKS and PP/NBR/PKS-PPMAH composites.

### CONCLUSIONS

Conclusively, tensile strength and Young's modulus decreased but elongation at break increased with increasing of NBR loading of both PP/NBR/PKS and PP/NBR/PKS compatibilized composites. Compatibilized composites show higher tensile properties. These findings were well supported by micrographs from the scanning electron microscopy. The FTIR spectrum of the PPMAH compatibilized indicates the C-O-C groups proven the bonding reaction in the PP/NBR matrices.

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