



**THE EFFECT OF COMPATIBILIZER AND  
COUPLING AGENT ON PROPERTIES AND  
CHARACTERIZATION OF ETHYLENE VINYL  
ACETATE/ NATURAL RUBBER/ POTASH  
FELDSPAR COMPOSITES**

by

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A thesis submitted in fulfillment of the requirements for the degree of  
Doctor of Philosophy

**School of Material Engineering  
UNIVERSITI MALAYSIA PERLIS**

**2017**

## ACKNOWLEDGEMENT

First at all, I would like to take this opportunity to thanks everyone who helped me in in the successful completion of this project.

The special thank goes to my helpful supervisor, Assoc. Prof. Dr Supri A. Ghani. The supervision, guidance, advice, patience, understanding and support that he gave truly help the progression and smoothness of my PhD project. Assoc. Prof. Dr Supri A. Ghani provided encouragement and motivations that helped me throughout my thesis-writing period. Besides, I would like to thank Dr Teh Pei Leng for her guidance and support.

Besides, I would like to thank Dean of School of Materials Engineering, Ass. Prof. Dr. Khairul Rafezi, and technicians Mr. Zaidi and Mr. Nasir who helped me in experimental, testing and analysis.

Last but not least, I would like to thanks to my family especially my parents who always there supporting me throughout my studies and to my fellow friends for their help and inspiration throughout this project and make it a success.

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## LIST OF ABBREVIATIONS

ATH	Alumina trihydrate
BU	Benzyl urea
COPE	Thermoplastic Copolyesters
DSC	Differential scanning calorimetry
DTG	Derivative thermogravimetric
EPDM	Ethylene propylene diene monomer
EPM-g-MA	Maleic anhydride grafted ethylene propylene rubber
EVA	Ethylene vinyl acetate
EVA-g-BU	Ethylene (vinyl acetate-grafted-benzyl urea)
EVA-g-PAH	Ethylene (vinyl acetate-grafted-phthalic anhydride)
FTIR	Fourier transform infrared spectroscopy
GA	Glycolic acid
GA-MA	Glycolic acid- maleic anhydride
HDPE	High density polyethylene
HIPS	High impact polystyrene
IA	Isophthalic Acid
IAMA	Isophthalic acid- maleic anhydride
LLDPE	Linear low density polyethylene
M100	Modulus at 100% elongation
MA	Maleic anhydride
MA-PP	Maleic anhydride modified polypropylene
NBR	Nitrile rubber
NR	Natural rubber



NR-g-MAH	Maleated natural rubber
NR-g-PDMMMP	Natural rubber grafted poly (dimethyl (methacryloyloxymethyl) phosphonate)
OENR	Oil extended NR
OMMT	Organo-montmorillonite
PAH	Phthalic Anhydride
PE-g-MAH	Polyethylene grafted maleic anhydride
PF	Potash Feldspar
PhHRJ-HDPE	Phenolic-modified polyethylene
Ph-PP	Phenolic modified polypropylene
PP	Polypropylene
PP-g-MAH	Maleic anhydride grafted polypropylene
PS	Polystyrene
PS-g-SBR	Styrene grafted styrene butadiene rubber copolymer
SBS	Styrenebutadiene-styrene block copolymer
SEM	Scanning electron microscope
SMR L	Standard Malaysian Rubber
TEM	Transmission electron microscopy
TG	Thermogravimetric
TPA	Thermoplastic Polyamide Elastomer
TPE	Thermoplastic elastomer
TPO	Thermoplastic polyolefin
TPU	Thermoplastic Polyurethanes Elastomers
WRP	Waste rubber powder

WTD	Waste tire dust
VA	Vinyl Acetate
XRD	X-Ray Diffraction

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## LIST OF SYMBOLS

$d$	Interparticle spacing
$\Delta H_f$	Enthalpy of fusion of the composite
$\Delta H_f^0$	Enthalpy of fusion
$I_1$	Intensity peak number 1
$I_2$	Intensity for peak number 2
Phr	Parts per hundred resin
$T_m$	Melting temperature
$T_{-50\% \text{ wt}}$	Temperature at 50 % weight loss
$W_1$	Weight of dry sample
$W_2$	Weight of wet sample

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## Sifat-sifat dan Pencirian Komposit Etilena Vinil Asetat / Getah Asli / *Potash Feldspar*

### ABSTRAK

Komposit etilena vinil asetat / getah asli / *potash feldspar* (EVA / NR / PF) telah dikaji. Komposit ini disediakan menggunakan Brabender Plasticoder pada suhu 160 °C dengan kelajuan rotor 50rpm. Kesan pembebanan PF, pelbagai jenis pengserasi, dan agen gandingan pada sifat mekanikal, sifat pembengkakan, sifat morfologi, sifat terma, pencirian spektroskopi inframerah, dan pencirian XRD terhadap komposit EVA / NR / PF telah dikaji. Dalam kajian ini, 70 phr etilena vinil asetat dan 30 phr getah asli telah digunakan sebagai polimer matrik. Sementara itu, *potash feldspar* dengan 5, 10, 15, 20 dan 25 phr telah digunakan sebagai bahan pengisi. Polietilena-dicantumkan-maleik anhidrida (PE-g-MAH), gabungan asid isofatolik dan anhidrida maleik (IAMA), etilena vinil asetat- dicantumkan- petalik anhidrida (EVA-g-PAH) and etilena vinil asetat-dicantumkan- benzil urea (EVA-g-BU) telah digunakan sebagai bahan pengserasi manakala agen gandingan silana dan asid gikolik- maleik anhidrida (GA-MA) digunakan untuk mengubahsuaikan permukaan PF. Penambahan PF untuk komposit EVA/NR telah mengurangkan kekuatan tegangan, pemanjangan pada takat putus, peratusan pembengkakan, dan jarak antara zarah tetapi telah meningkatkan modulus pada 100% pemanjangan (M100) dan peratusan penghabluran. Morfologi permukaan tegangan-patah telah menunjukkan penumpuan PF apabila peningkatan jumlah PF. Apabila jumlah PF meningkat, kestabilan terma untuk komposit didapati meningkat sebab suhu penguraian dan sisa jisim adalah tinggi pada jumlah PF yang tinggi. Komposit dengan pengserasi PE-g-MAH, IAMA, EVA-g-PAH, and EVA-g-BU masing-masing menunjukkan purata kenaikan sebanyak 3.19%, 4.72%, 11.24%, dan 21.39% di dalam kekuatan tegangan; 5.52%, 23.69%, 38.49%, dan 21.25% di dalam M100 berbanding dengan komposit EVA/NR/PF. Walau bagaimanapun, penyusutan sebanyak 3.97%, 5.51%, 6.24%, and 3.02% dalam pemanjangan pada waktu putus, dan pengurangan sebanyak 8.10%, 3.18%, 3.37%, dan 3.66% dalam peratusan pembengkakan telah dilaporkan untuk komposit dengan pengserasi PE-g-MAH, IAMA, EVA-g-PAH, dan EVA-g-BU berbanding dengan EVA / NR / PF komposit. Komposit dengan pengserasi PE-g-MAH, IAMA, EVA-g-PAH, dan EVA-g-BU mempamerkan peningkatan dalam kestabilan terma tetapi penyusutan untuk peratusan penghabluran, dan jarak antara zarah berbanding dengan komposit EVA / NR / PF. PF telah diubahsuaikan dengan agen gandingan silana dan GA-MA di dalam etanol. Peningkatan 10.78% dan 13.66% untuk kekuatan tegangan manakala 77.79% dan 14.68% untuk M100 telah dilaporkan di komposit EVA/NR/PF<sub>Silane</sub> dan EVA/NR/PF<sub>GA-MA</sub> manakala pemanjangan pada waktu putus dan peratusan pembengkakan telah menurun. Komposit EVA/NR/PF<sub>Silane</sub> dan EVA/NR/PF<sub>GA-MA</sub> telah dipamerkan peratusan penghabluran dan jarak antara zarah yang lebih rendah tetapi kestabilan terma yang lebih tinggi berbanding dengan komposit EVA / NR / PF. SEM morfologi bagi semua komposit telah dipamerkan permukaan yang kasar dan telah menunjukkan penambahan untuk lekatan antara muka.

## Properties and Characterization of Ethylene Vinyl Acetate/ Natural Rubber/ Potash Feldspar Composites

### ABSTRACT

The composites of ethylene vinyl acetate/ natural rubber/ potash feldspar (EVA/NR/PF) were studied. The composites were prepared by using Brabender Plasticoder at 160°C with 50rpm of the rotor speed. The effect of PF loading, various types of compatibilizers, and coupling agents on mechanical properties, swelling behaviour, morphology properties, thermal properties, spectroscopy infrared, and XRD characterization of the composites were investigated. In this study, 70 phr of ethylene vinyl acetate and 30 phr of natural rubber were used as the polymer matrix. Meanwhile, potash feldspar with 5, 10, 15, 20, and 25 phr was used as filler. Polyethylene-grafted-maleic anhydride (PE-g-MAH), the combinations of isophthalic acid and maleic anhydride (IAMA), ethylene vinyl acetate- grafted- phthalic anhydride (EVA-g-PAH), and ethylene vinyl acetate- grafted- benzyl urea (EVA-g-BU) were used as compatibilizer while silane coupling agent and glycolic acid- maleic anhydride (GA-MA) were used to modify the surface of PF. The addition of PF into EVA/NR composites has reduced the tensile strength, elongation at break, percentage mass swell, and interparticle spacing but improved modulus at 100% elongation (M100) and percentage of crystallinity. The tensile fractured surface morphology had illustrated agglomerations of PF at higher PF loading. At higher PF loading, the thermal stability of the composites was found higher as temperature of decomposition and residual were higher at higher PF loading. The compatibilized composites with PE-g-MAH, IAMA, EVA-g-PAH, and EVA-g-BU showed an average of 3.19%, 4.72%, 11.24%, and 21.39% respectively higher in tensile strength; 5.52%, 23.69%, 38.49%, 21.25% respectively higher in M100 than EVA/NR/PF composites. However, 3.97%, 5.51%, 6.24%, and 3.02% lower in elongation at break and 8.10%, 3.18%, 3.37%, and 3.66% lower in mass swell percentage had been reported in PE-g-MAH, IAMA, EVA-g-PAH, and EVA-g-BU compatibilized composites respectively compared to EVA/NR/PF composites. Besides, the compatibilized composites presented higher thermal stability but lower percentage of crystallinity, and interparticle spacing compared to EVA/NR/PF composites. PF was treated with silane coupling agents and GA-MA in ethanol. The increment of an average of 10.78% and 13.66% in tensile strength and 77.79% and 14.68%, in M100 were reported in EVA/NR/PF<sub>Silane</sub> and EVA/NR/PF<sub>GA-MA</sub> composites respectively while the elongation at break and mass swell were decreased for the composites. EVA/NR/PF<sub>Silane</sub> and EVA/NR/PF<sub>GA-MA</sub> composites exhibited lower percentage of crystallinity and interparticle spacing but higher thermal stability in comparison to EVA/NR/PF composites. The SEM morphology for all the compatibilized and treated composites exhibited rough surface and indicated that the interfacial adhesion had been improved.

# CHAPTER 1

## INTRODUCTION

### 1.1 Research Background

Polymer composites are defined as materials consisting two or more distinct phases that their separate identities were retained. In polymer composites, matrix phase is primary phases having continuous character whereas dispersed phases is secondary phase and usually stronger than matrix (Thomas, 2012). Polymer composite materials had replaced many conventional materials in many applications. This is due to they are more design flexibility and corrosion resistance compare with other conventional composites (Leng et al., 2010). Polymer composites received attention in structural engineering due to their unique properties such as reduced product cost and good mechanical properties.. These materials were applied in high- performance, light weight applications such as motor sport and aerospace (Greenhalgh, 2009).

Thermoplastic elastomers (TPEs) are elastomeric materials that can be processed as a thermoplastic and can be recycled. Besides, TPO do not need a complex system of chemicals for crosslinking. TPEs have the combinations processing properties of thermoplastic materials at elevated temperature and characteristics of vulcanized elastomers at service and room temperatures. TPE can be reprocessed and have economic advantages (Pechurai et al., 2008). The plastic matrix which is continuous provides the melt processing and the dispersed rubber phase provides the elasticity to the blend after deformation (Chang et al., 2006).

Many researches had been done on the properties of TPEs. Dynamic mechanical and thermal properties of a chemically modified polypropylene/natural rubber blend had

been observed by Benmesli et al. (2014). In the study, they found out that NR-g-MAH/PP-g-MAH blend had lower loss modulus than unmodified NR/PP blend, reflected the interfacial interaction that resulted from the chemical modification. Others, the increment of 5 °C in glass transition temperature in NR-g-MAH/PP-g-MAH blend was observed. They claimed that the improvement in the interactions was due to the loss of the  $\beta$  transition of PP phase in the NR-g-MAH/PP-g-MAH blend.

Thermal and crystallization behaviour of isotactic polypropylene/nitrile rubber blends had been reported by George et al. (2000). They reported that the dipolar interaction between polar group of NBR and maleic anhydride groups of PP enhanced the decomposition temperature in the PP/NBR blend with the incorporation of MA-PP or Ph-PP as compatibilizers in the blend.

The morphology and mechanical properties of polystyrene (PS) and natural rubber (NR) blend had been studied by Asaletha et al. (1999). Two phase structure had been shown in morphology. The dispersed phase in 30/70 NR/PS blend is NR and PS is the continuous phase whereas polystyrene formed the dispersed phase and NR formed the continuous phase in 70/30 NR/PS blend. With the increase in rubber content, tensile strength and tear strength decreased whereas impact strength increased.

Ethylene vinyl acetate (EVA) consists of random copolymer of vinyl acetate and ethylene, which had been used in many applications (Moly et al., 2007; Thaworn et al., 2012). EVA can be used as elastomer, thermoplastic and thermoplastic elastomer depending on the vinyl acetate (VA) content in EVA copolymer (Ma et al., 2014; Stelescu et al., 2012). Besides, the vinyl acetate content varied the properties of EVA. The polar VA group can compatible with polar polymer and fillers (Hosier et al., 2010). EVA has excellent impact resilience, optical properties and flexibility. EVA is largely applied in corrosion protection, electrical insulation, electrical cable sheathing,

packaging of components and shoe industry due to its easy acceptance of additives, low density, resistance in colour change, low cost and physic-chemical properties (Bidsorkhi et al., 2015; Jin et al., 2007; Park et al., 2005).

Natural rubber (NR) is one of the common elastomer used as polymer matrix in thermoplastic elastomer due to its mechanical properties and unique strain crystallization (Liu et al., 2014). NR performs high tensile strength, good resilience, low compression set, tear and wear resistance, and good electrical properties. NR is chemically resistant to alkalis, acids and alcohol (Visakh et al., 2013). NR is widely used in automotive tires, tire tread, and mechanical goods (Harper et al., 2003). Natural rubber is applied in over 40 000 products such as medical devices, surgical gloves, and automobile tires (Nanthini et al., 2016).

EVA and NR blend had gain attention either in vulcanized or unvulcanised. Many attractive properties of NR had made it suitable to blend with EVA. Intharapat et al. (2013) had studied the compatibilization of NR/EVA blends in the existennce of NR-g-PDMMMP (natural rubber grafted poly (dimethyl (methacryloyloxymethyl) phosphonate)) as compatibilizer. They reported that compatibilization effect was the best at 7 wt.% of compatibilizer. Besides that, finest morphology of NR/EVA blend with NR-g-PDMMMP can be observed.

Sujith et al. (2005) had studied the barrier properties of natural rubber/ ethylene vinyl acetate/ carbon black composites. They reported that the blending of EVA and NR had reduced the solvent sensitivity of NR due to the introduction of rigid EVA regions into NR. Besides that, molecular sorption by heterogeneous natural rubber/ poly (ethylene-co-vinyl acetate) blend systems was studied by Sujith et al. (2006). They claimed that the solvent uptake for higher EVA content was lower. The vulcanization of NR/EVA using sulphur exhibited the highest solvent uptake followed by the mixed