



**UNIVERSITI PUTRA MALAYSIA**

**PREPARATION AND CHARACTERIZATION OF  
POLYPROPYLENE-NATURAL RUBBER /CLAY NANOCOMPOSITES**

**GOH WEI CHEE**

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**PREPARATION AND CHARACTERIZATION OF  
POLYPROPYLENE-NATURAL RUBBER /CLAY NANOCOMPOSITES**

**By**

**GOH WEI CHEE**

**Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia,  
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**Chairman: Professor Wan Md. Zin bin Wan Yunus, Ph.D.**

**Faculty: Science**

In this study, octadecylamine (ODA-MMT) and dodecylamine (DDA-MMT) modified natrium-montmorillonites (Na-MMTs) were used to prepare polypropylene-natural rubber /clay nanocomposites. The products were characterized using Fourier Transform Infrared (FTIR) spectroscopy, X-ray diffraction (XRD), Energy Filtering Transmission Electron Microscopy (EFTEM), Thermogravimetric Analyser (TGA) and tensile measurement. To prepare the nanocomposites, the ODA-MMT and DDA-MMT were blend with polypropylene and natural rubber in the ratio of 70: 30, by using an internal mixer. The study of blending speed, temperature and period shows that under the following blending conditions: 80 r.p.m. of rotor speed, temperature of 180°C and 6 minutes of blending period, the hybrid produced with maximum tensile strength and elongation at break was obtained. The optimum clay content to give maximum tensile strength is 1 php. In order to improve the mechanical properties of the blend, different



amount of *N, N'*-m- phenylenedimaleimide (HVA-2) was blended with PPNR and their properties were also studied by tensile measurement, TGA and dynamic mechanical analysis (DMA). One php of HVA-2 was taken as the optimum concentration to be incorporated in PPNR because it gave the highest tensile improvement. The tensile properties and thermal degradation behaviour of PPNR/clay crosslinked with HVA-2 were then compared with the properties of PPNR/clay which consists of maleic anhydride grafted polypropylene (MAGPP) as a compatibilizer. HVA-2 crosslinked PPNR/clay shows higher tensile strength and elongation at break improvement compared to that of the MAGPP compatibilized PPNR/clay. However, the TGA thermograms of these two systems are very similar. The PPNR/HVA-2/clay nanocomposites were further modified by changing their clay contents. XRD and TEM analyses show that octadecylamine modified montmorillonite (ODA-MMT) and dodecylamine modified montmorillonite (DDA-MMT) produce a mixture of intercalation and flocculation, and of intercalation and exfoliation in the hybrids, respectively. By incorporating 1 and 2 php of ODAMMT into the HVA-2 crosslinked PPNR, the tensile strength of the hybrid increases about 10%. One php of DDAMMT clay loaded in the blend causes 7% of increment in tensile strength. The thermal degradation of both organoclay loaded blends is almost the same as the sample with the low clay content. Increase the modified clay content to 5 php improves the degradation temperature of 20°C.



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**PENYEDIAAN DAN PENCIRIAN  
NANOKOMPOSIT POLIPROPILENA-GETAH ASLI/TANAH LIAT**

Oleh

**GOH WEI CHEE**

**Disember 2006**

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Dalam kajian ini, natrium montmorillonite (NaMMT) yang telah diubahsuai oleh oktadesilammonium (ODA-MMT) dan dodesilammonium (DDA-MMT) telah digunakan untuk menyediakan nanokomposit polipropilena-getah/tanah liat. Pencirian komposit yang dihasilkan dibuat dengan menggunakan kaedah spektroskopi inframerah (FTIR)s, analisis pembelauan sinar-X (XRD), mikroskopi elektron penyebaran (EFTEM), penganalisis termogravimetri (TGA), dan pengukuran tegangan. Untuk menyediakan nanokomposit, tanah liat yang diubahsuai oleh oktadesilammonium dan dodesilammonium diadun dengan polipropilena (PP) dan getah asli (NR) dalam nisbah 70: 30, dengan menggunakan pencampur dalaman. Kajian bagi menentukan jangka masa adunan, kadar adunan dan suhu adunan menunjukkan bahawa pada keadaan adunan berikut: kadar pusingan roda 80 rpm, suhu adunan 80°C dan jangka masa selama 6 minit, adunan yang dihasilkan mempunyai kekuatan tegangan dan pemanjangan pada



takat putus yang maksima. Kandungan tanah liat yang optimum untuk adunan mencapai kekuatan tengangan maksima ialah 1 php. Untuk mempertingkatkan sifat mekanik adunan, kandungan *N, N'*-m- fenilindimaleimide (HVA-2) yang berbeza telah ditambah ke dalam PPNR dan sifat-sifat adunan dikaji dengan pengukuran tegangan, TGA dan analisis mekanika dinamik (DMA). Satu php HVA-2 telah diambil sebagai kandungan optimum untuk dicampurkan ke dalam PPNR kerana ia memberi peningkatan tengangan yang tertinggi. Sifat tegangan dan perosotan terma bagi adunan PPNR/tanah liat yang disambung silang dengan HVA-2 kemudian dibandingkan dengan sifat-sifat adunan PPNR/tanah liat yang mengandungi polipropilena dicangkukkan malik anhidrida (MAgPP) sebagai agen penserasi. Adunan PPNR/tanah liat yang disambung silang dengan HVA-2 menunjukkan peningkatan kekuatan tegangan dan pemanjangan pada takat putus yang tinggi jika dibanding dengan PPNR/tanah liat yang diserasi oleh MAgPP. Akan tetapi, termogram TGA bagi kedua-dua sistem lebih kurang sama. Kajian bagi PPNR/HVA-2/tanah liat nanokomposit dilanjutkan dengan mencampurkan kandungan tanah liat yang berlainan di dalam adunan. Analisis XRD dan TEM menunjukkan bahawa ODA-MMT dan DDA-MMT masing-masing menghasilkan adunan jenis interkalasi dan pengumpulan serta interkalasi dan kelupas. Dengan mencampurkan 1 dan 2 php ODA-MMT ke dalam adunan PPNR/tanah liat yang disambung silang oleh HVA-2, kekuatan tegangan bagi adunan tersebut diperhatikan meningkat lebih kurang 10%. Pencampuran 1 php DDA-MMT dalam adunan menyebabkan peningkatan kekuatan tegangan sebanyak 7%. Perosotan terma bagi adunan yang mengandungi kedua-dua jenis tanah liat terubah adalah lebih kurang sama pada kandungan tanah liat yang rendah. Peningkatan kandungan tanah liat terubah kepada 5 php telah menyebabkan peningkatan perosotan terma sebanyak 20°C.

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I certify that an Examination Committee has met on 11 June 2007 to conduct the final examination of Goh Wei Chee on her Master of Science thesis entitled "Preparation and Characterization of Polypropylene-Natural Rubber /Clay Nanocomposites" in accordance with Universiti Pertanian Malaysia (Higher Degree) Act 1980 and Universiti Pertanian Malaysia (Higher Degree) Regulations 1981. The Committee recommends that the candidate be awarded the relevant degree. Members of the Examination Committee are as follows:

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## DECLARATION

I hereby declare that the thesis is based on my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at UPM or other institutions.

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## LIST OF ABBREVIATIONS

CEC	Cation exchange capacity
DDA	Dodecylamine
DDA-MMT	Dodecylamine modified montmorillonite
DMA	Dynamic mechanical analysis
DTG	Derivative thermal gravimetry
EPDM	Ethylene-propylenediene rubber
EPM	Ethylene-propylene rubber
FTIR	Fourier transform infrared
HVA-2	N, N'-m-phenylenedimaleimide (HVA-2)
LLDPE	Low density polyethylene
MAGPP	Maleic anhydride grafted polypropylene
Na-MMT	Sodium montmorillonite
NBR	Nitrile rubber
NR	Natural rubber
ODA	Octadecylamine
ODA-MMT	Octadecylamine modified montmorillonite
php	Part per hundred polymer
PP	Polypropylene
PPNR	Polypropylene-natural rubber
SBS	styrene-butadiene-styrene
SIS	styrene-isoprene-styrene



SMR CV	Standard Malaysian Rubber constant viscosity
TEM	Transmission electron microscopy
T <sub>g</sub>	Glass transition temperature
TGA	Thermogravimetic analysis
T <sub>max</sub>	Temperature at maximum decomposition
TPE	Thermoplastic elastomer
TPO	Thermoplastic olefin
TPV	Thermoplastic vulcanizate
XRD	X-ray diffraction



## CHAPTER I

### INTRODUCTION

Polymer blend is a kind of material which consists of two or more polymers or copolymers. The blending of different types of polymers is aim to get a material with the properties in between the different types of polymers. Commercial polyblends are commonly of three types: (i) elastomer-elastomer blends, (ii) thermoplastic-thermoplastic blends and (iii) thermoplastic-elastomer blends (Ghosh, 2002). Among them, thermoplastic elastomer or rubber modified thermoplastic (depends on the composition of the blend) had gained much industrial interest. The blending of plastic and rubber offer the advantages of elastomer behaviour at room temperature and thermoplastic behaviour at processing temperature (Kuriakose *et al.* 1984; Montoya *et al.* 2004). They bridge the gap between rubber and plastic in both processing and final properties. During the period from 1960 to 1975, a large number of patents on elastomer-thermoplastics blends, especially those on ethylene-propylene rubber (EPM), ethylene-propylenediene rubber (EPDM) and polypropylene (PP) were issued (Fisher, 1972; Fritz and Shih, 1973; Bodungen and Meredith, 1974).

The automotive industry is increasingly using flexible plastics for soft front and rear ends, side protection strips, bumpers and interior trim. These components are designed to be several orders of magnitude more flexible than steel, yet stiff enough for components to support their own weight. Hence dimensional stability at elevated temperatures is required, which includes the effect or exposure to sunlight and, in the



case of painted parts, to a baking temperature of at least 120°C (Elliot, 1990).

In order to improve the toughness and stiffness of the materials, to enhance their barrier properties, to enhance their resistance to fire, or simply to reduce cost, manufacturers fill polymers with particles. However, due to the size of the fillers, large loading are required to significantly increase stiffness, which can result in poor processability, lower ductility, and a rough surface finish. By replacing such fillers with layered silicates could potentially alleviate these weaknesses (Lee *et al.*, 2005).

Montmorillonite is one of the common natural clay from smectite family. The silicate layer consists of two fused silica tetrahedral sheets sandwiching an edge-shared octahedral sheet of either aluminium or magnesium hydroxide.  $\text{Ca}^+$  and  $\text{Na}^+$  situated between the layers to counterbalance the negative charges. Due to the high hydrophilicity of the clay, water molecules are usually present between the layers, causing low surface activity in between the clay and matrix. Therefore the clay particles could only be dispersed on the microscale in the polymer matrix and thus give the properties of conventional composites (Kornmann, 1999).

Ion-exchange process is well established in exchanging the hydrophilic clay into organophilic. Interlayer cations can be exchanged with alkyl ammonium or alkylphosphonium in order to give a polar surface. Polymers were found easier to penetrate such modified clay. The insertion of polymer matrix into the nanometer scale silicate layer further increase the gap between the silicate layers and forms an intercalated nanocomposite (Ray and Okamoto, 2003). Depending on the ratio and





nature of the components and also the method using, delamination of silicate layers can occur and exfoliate polymer-clay nanocomposite can be obtained. Without the registry between the silicate layers, exfoliate polymer-clay nanocomposites were reported had gained significant improvement in stiffness, strength, heat resistance and barrier properties if compared with its unfilled counterpart.

A number of investigations on rubber-clay nanocomposites (Usuki 2002; Arroyo *et al.*, 2003; Zheng *et al.*, 2004) as well as polypropylene-clay nanocomposites (Liu and Wu 2001; Nam. *et al.*, 2001; Garcia-Lopez. *et al.*, 2003; Xu. *et al.*, 2002; Zheng. *et al.*, 2004; Morgan and Harris 2003; Tang *et al.*, 2003; Nowacki. *et al.*, 2004; Wang. *et al.*, 2004; Zhang. *et al.*, 2004) have been carried out. However, only a few researches were published on the polypropylene-elastomer-clay blend nanocomposites (Mishra *et al.*, 2004; Lee *et al.*, 2005; Lee *et al.*, 2006; Thompson and Yeung, 2006). Apparently, no research was published on polypropylene-natural rubber-clay blend nanocomposites.

## **LITERATURE REVIEW**

### **Polymer blends**

Polymer blends are available either in homogeneous or heterogeneous (Ghosh, 2002). In other words, they can be classified as miscible, partially or nearly miscible or immiscible blend. Miscible blends refer to those blends that are homogenous at a molecular level while those blends that are homogenous at some temperatures and

