



**PROPERTIES OF ETHYLENE PROPYLENE
DIENE MONOMER BLENDED WITH RECYCLED
ACRYLONITRILE BUTADIENE RUBBER FILLED
BAMBOO PARTICULATE (EPDM/rNBR)**

by

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LIST OF ABBREVIATIONS

ASTM	American society for testing and materials
ENR	Epoxidised natural rubber
EPDM	Ethylene propylene diene monomer
ESBS	Styrene – epoxidised butadiene – styrene triblock copolymer
FTIR	Fourier transform infrared
IRHD	International Rubber Hardness Degree
NBR	Acrylonitrile butadiene rubber
NR	Natural rubber
PFMs	Polyfunctional monomers
phr	Part per hundred rubber
rNBR	Recycled acrylonitrile butadiene rubber
rPA	recycled polyamide
SBR	Styrene butadiene rubber
SEM	Scanning electron microscopy
TGA	Thermogravimetric analysis
TMTD	Tetramethyl thiuram disulfide
TOR	Trans Polyoctylene Rubber
vNBR	Virgin nitrile butadiene rubber
WRP	Waste rubber powder
WTD	Waste tire dust

LIST OF SYMBOLS

cm	Centimeter
E_b	Elongation at break
Kg/m^3	Relative density
M100	Stress at 100 % elongation
M_c	Molecular weight between crosslinks
M_H	Maximum torque
M_L	Minimum torque
mL	Milliliter
mm	millimeter
MPa	MegaPascal
Q_m	Total weight swelling
t_2	Scorch time
t_{90}	Cure time
v	Degree of crosslinking density
V_r	Volume fraction of the swollen rubber
V_s	Molar volume of the solvent
χ	Interaction parameter of rubber
θ_1	Initial angle (45°)
θ_2	Maximum rebound angle
μm	Micrometer

Sifat-sifat Etilena Propilena Diena Monomer Diadun dengan Getah Akrilonitril Butadiena Kitar Semula Dipenuhi Zarahhan Buluh (EPDM/rNBR)

ABSTRAK

Pengadunan getah telah dipilih sebagai salah satu teknik dan peluang untuk mengatasi isu-isu alam sekitar berkenaan bahan buangan getah. Getah etilena propilena diena monomer (EPDM) diadun dengan getah akrilonitril butadiena kitar semula (rNBR) dengan cara mengadun bersama boleh menghasilkan bahan polimer baru. Sifat-sifat rNBR dan buluh yang digunakan sebagai bahan pengisi dikaji dengan menggunakan Ujian Analisis Spektroskopi Inframerah (FTIR), Ujian Analisis Termal Gravimetrik (TGA), dan Analisis Mikroskop Penskanan Elektron (SEM) ke atas permukaan. Siri 1 mengkaji campuran EPDM diadun dengan kitar semula NBR pada saiz dan beban yang berbeza. Keputusan menunjukkan bahawa adunan EPDM/rNBR pada 15 phr rNBR yang bersaiz paling kecil (S1: 250-500 μm) mempunyai masa skorch, t_2 yang lebih rendah, dan maksimum tork yang lebih tinggi, M_H berbanding dengan rNBR bersaiz besar (S2: 5-10 cm). Sifat-sifat tegangan EPDM/rNBR (S1) menunjukkan kesan sinergi sehingga 15 phr rNBR di mana ia mempunyai nilai yang lebih tinggi daripada kekuatan tegangan dan ketumpatan sambung silang. Siri 2 menyiasat kesan buluh sebagai bahan pengisi dalam EPDM/rNBR. Tiga saiz pengisi buluh telah digunakan dan penambahan pengisi buluh pada saiz yang kecil mengisi adunan EPDM/rNBR (S1: $\leq 125 \mu\text{m}$) pada kandungan 35 phr pengisi buluh menunjukkan sifat lebih baik dari segi ciri-ciri matang, kekuatan tegangan dan ciri-ciri fizikal berbanding dengan adunan EPDM/rNBR yang dipenuhi pengisi buluh bersaiz besar (S2: 125 - 250 μm dan S3: 250 -500 μm). Pengisi buluh yang bersaiz kecil bertindak sebagai bahan pengisi lebih baik dalam adunan EPDM/rNBR. Siri 3 mengkaji penambahan penserasi getah trans-polioktilena (TOR) ke dalam adunan EPDM/rNBR. Penserasi TOR memberikan pertambahan baik interaksi antara muka iaitu antara rNBR dan EPDM matriks, sekali gus meningkatkan keserasian adunan. Penserasi, TOR telah menjadi sebahagian daripada rangkaian. Walau bagaimanapun, penambahan beban TOR secara berlebihan telah menyekat pergerakan molekul yang menghadkan orientasi rantai rangkaian. Secara keseluruhan, sifat matang, tegangan dan fizikal menunjukkan peningkatan pada 2 phr beban TOR dan berbanding nisbah gabungan lain.

Properties of Ethylene Propylene Diene Monomer Blended with Recycled Acrylonitrile Butadiene Rubber Filled Bamboo Particulate (EPDM/rNBR)

ABSTRACT

Rubber blending has been selected as one of the techniques and opportunity to overcome the environmental issues regarding rubber waste product. The ethylene propylene diene monomer (EPDM) rubber blended with recycled acrylonitrile butadiene rubber (rNBR) by means of blending together can produce new polymeric material. The properties of rNBR and bamboo fillers used as fillers were characterized using Fourier Transform Infrared Radiation (FTIR), Thermal Gravimetric Analysis (TGA) and Scanning Electron Microscopy (SEM) on the tensile fracture surfaces. Series 1 studied the blending of EPDM blended with recycled NBR at different sizes and loadings of rNBR. Results indicated that EPDM/rNBR blends at 15 phr rNBR with smallest size (S1: 250 – 500 μm) having lower scorch time, t_2 and higher maximum torque, M_H compared to large size (S2: 5 – 10 cm). The tensile properties of EPDM/rNBR (S1) showed a synergism effect up to 15 phr rNBR where it has higher value of tensile strength and crosslink density. Series 2 investigated the effects of bamboo filled EPDM/rNBR. Three different sizes of bamboo fillers has been used and the addition of smallest size bamboo fillers filled EPDM/rNBR blends (S1: $\leq 125 \mu\text{m}$) at 35 phr bamboo fillers content showed better properties in terms of curing properties, tensile strength and physical properties in comparison with EPDM/rNBR blends filled large size bamboo fillers (S2: 125 – 250 μm and S3: 250 -500 μm). Bamboo fillers with smallest size act as better reinforcing filler in EPDM/rNBR blends. Series 3 studied the addition of compatibilizer, trans-polyoctylene rubber (TOR) into EPDM/rNBR blend. Compatibilizer TOR improved the interfacial interaction between rNBR and EPDM matrix, thus improving the compatibility of the blends. TOR has become a part of the network. However, the excessive increment of TOR loading restricts the molecular mobility which limited the orientation of the network chain. The overall curing properties, tensile and physical properties showed an enhancement at 2 phr TOR loading rather than other blend ratios.

CHAPTER 1

INTRODUCTION

1.1 Recycling of Rubber Waste

Currently, Malaysia Rubber Board on Natural Rubber Statistics 2015 reported that world rubber production of natural and synthetic rubber dropped drastically in 2015 compared to the previous year. Figure 1.1 shows that the world rubber production on 2000 until 2015. The rubber production was about 17 681 tonnes in year 2000 and continue to increase till 2013 but decrease highly to 19 652 tonnes in year 2015 respectively. The decrease was due to the world less consumption of rubber and consequently Malaysia's rubber production also dropped respectively. Figure 1.2 shows a pattern of world rubber consumption of natural and synthetic rubber. The rubber consumption decreased about 20 111 tonnes in 2015 reported by International Rubber Study Group (IRSG) ("Natural Rubber Statistics ").The decrement of the world rubber production and consumption can be due to the high cost of the rubber based product. Hence, some initiatives taken such as recycling reclaim and reused have been given a big attention. This tend to give beneficial to people yet supporting in reduce environmental issues.

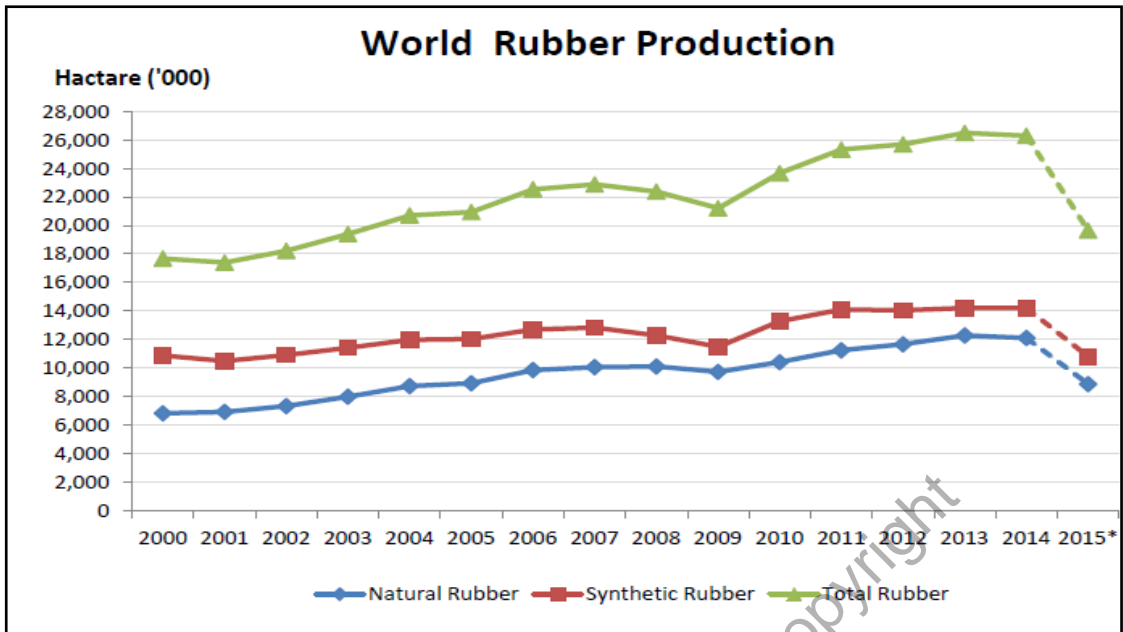


Figure 1.1: World rubber production year 2000-2015 ("Natural Rubber Statistics ")

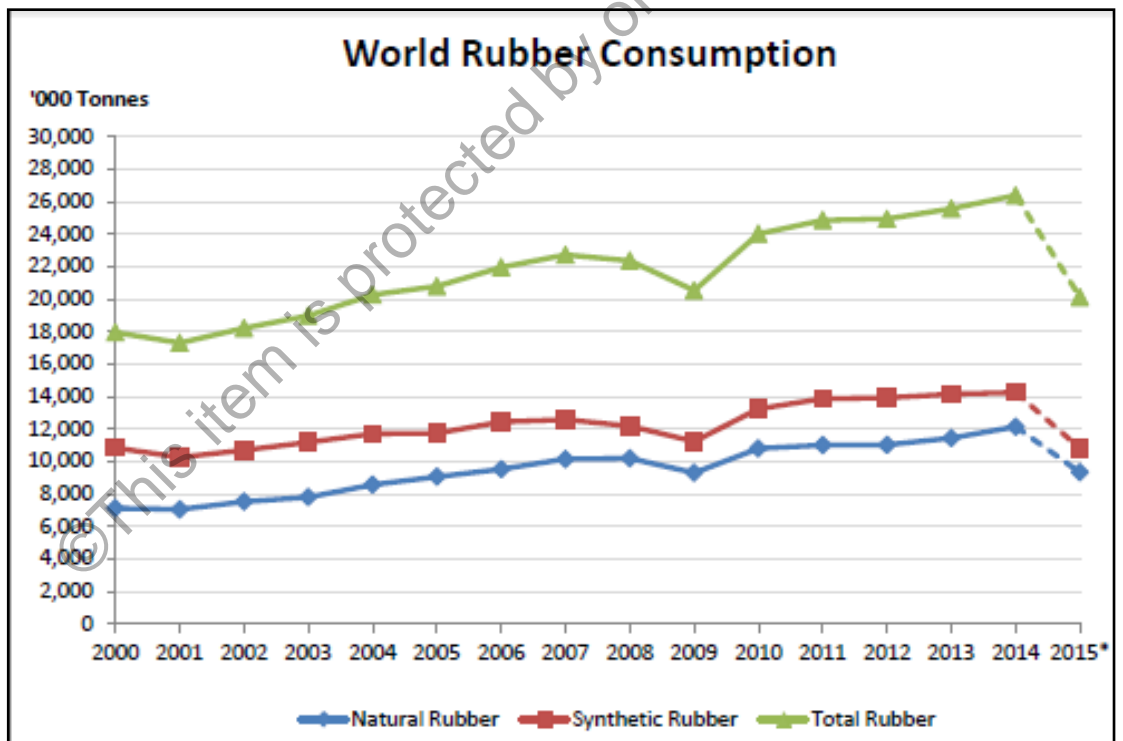


Figure 1.2: World rubber consumption year 2000 - 2015 ("Natural Rubber Statistics ")

The term recycle is best described as a process of changing waste materials into new products to prevent waste of potentially useful materials. Recycled rubber can be categorized into reclaimed rubber, ground rubber or reprocessed synthetic rubber. Recycling technique reduces the consumption of fresh raw materials, reduce energy usage and reduce environmental pollution such as air and water pollution. Recyclable waste materials especially recycled rubber does not decompose easily due to its crosslinked structure and the presence of stabilizers and other additives (Noriman et al., 2012). Polymer blend such as elastomers together with recycled waste has been applied to meet the performance and processing requirements to manufacture a wide range of variety rubber based products such as playground surfaces, recycled rubber flooring, adhesives glues, sporting mats, floats, marine and automotive parts (Noriman and Ismail, 2011).

Malaysia is the world global supplier of latex products mainly glove, condom and catheters. Hence, there was a great interest in polymer industry regarding to the development of cost effective techniques converting waste and used rubber into potential processable form (Noriman et al., 2008). Figure 1.3 expresses the distribution of Malaysia rubber product companies by sector in 2015. Among all of the rubber products, glove product leads the sector where 54 companies involved in glove production. This huge number approximately generated a high amount of rejected rubber glove during the production line. Therefore, the practice of rubber recycling was introduced and expanded for society awareness in green issues as well as saving resources. Blending is one of the easy method and cost effective way to produce a new combination properties of recycled rubber waste. The addition of recycled rubber waste in form of ground waste vulcanizate or reclaim improve the physical and mechanical

properties as well as modify processing characteristics and lower the cost of final product. In some blends, there is a difficulty to achieve the compatibility between the rubbers. This was due to the blends characteristics which depend on type of rubber, formulations and cure conditions. Previously, a number of possible applications of various forms or rubber in broad disciplines have been studied and reported. Eventhough many research regarding the potential of using rNBR in rubber blend were carried out, the study of ethylene propylene diene monomer (EPDM) blended with recycled nitrile butadiene rubber (rNBR) has not been discovered.

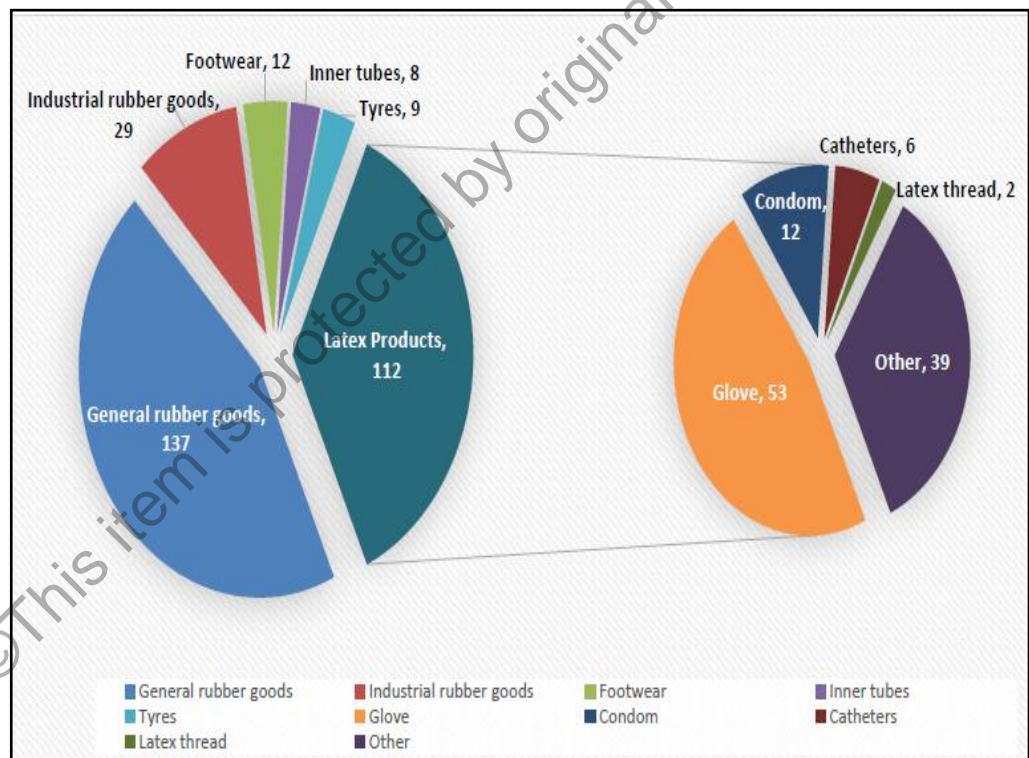


Figure 1.3: Distribution of Malaysia Rubber Product Companies by Product Sector ("Natural Rubber Statistics ")

1.2 Problem statement

Recently, the waste materials generate serious environmental issues. The recycle and reduce scrap waste, the disposal of rubber waste products is of significant concern. Landfill disposal and transportation costs are expected to increase. Considering its economic and environmental advantages, recycling is one of the best options. The waste materials such as glove were recycled and used in rubber industry as a value added. Most of recycled material such as recycled tyres and other consumer product have dangerous chemical and harmful to be use in longer period during exposure. The recycled material need to undergo certain processes such as hygenic process and this caused the recycling method is sometimes very expensive.

Based on economical issues on global world demand products, rubber has been used to replace metal, wood and etc. In order to increase the quality of the rubber based products, many filler from either natural or synthetic has been used to enhance the properties of the product. Since bamboo is natural filler which provide significance properties, the uses of bamboo as filler in rubber blends is being study to see whether this filler can improve and enhanced the mechanical properties of the rubber blends.

Blending of rubber can either be compatible or incompatible depends on many factors. Some of the factors involve the types of rubber use, formulation, processing and cure conditions. In order to achieve the compatibility of the rubber blending that differs in polarity, the use of the third material in rubber blends such as compatibilizer, bonding agent, coupling agent and homogenizing agent were introduced.

In this study, ethylene propylene diene monomer (EPDM) blended with recycled acrylonitrile butadiene rubber (rNBR) at different blend formulations were observed and analyzes to see whether it may enhance the properties of the vulcanizate. The addition of bamboo fillers and compatibilizer in EPDM/rNBR were studied. Although there are many recent studies of rubber blend using rNBR but none of them focuses on rNBR blend with EPDM rubber.

1.3 Research objectives

The main objectives of this study were to review the effects of recycled NBR glove (rNBR) as a part in ethylene propylene diene monomer rubber blends (EPDM/rNBR) on curing properties, tensile, physical and morphology studies. Therefore, it can be outline as follows:

- a. To characterize the properties of recycled NBR gloves (rNBR) and bamboo fillers using particle size analyzer, Fourier Transform Infrared Radiation (FTIR), Thermal Gravimetric Analysis (TGA) and Scanning Electron Microscopy (SEM) on the surface.
- b. To determine the effects of different size of recycled acrylonitrile butadiene rubber and its blend ratio on properties of ethylene propylene diene monomer/recycled acrylonitrile butadiene rubber (EPDM/rNBR) blends.

- c. To examine the effects of different sizes bamboo fillers and its blend ratio filled ethylene propylene diene monomer/recycled acrylonitrile butadiene rubber (EPDM/rNBR) blends.
- d. To investigate the effects of compatibilizer, trans-polyoctylene rubber (TOR) content on the properties of bamboo filled ethylene propylene diene monomer/recycled acrylonitrile butadiene rubber (EPDM/rNBR) composites.

1.4 Scope of study

Characterization and properties of recycled acrylonitrile butadiene rubber (rNBR) and bamboo fillers were analyzed by using FTIR and SEM on the surfaces. The ethylene propylene-diene monomer rubber (EPDM) blended with different size recycled NBR (rNBR) were studied at different formulation and blend ratio (95/5, 85/15, 75/25, 65/35 phr). The optimum blend ratio of EPDM/rNBR blends was then further studied by adding bamboo fillers as reinforcement. Bamboo fillers used in EPDM/rNBR blends were examined at 3 different sizes (≤ 125 , 125-250, and 250-500 μm) with different formulations and blend ratios. Then, the best result obtained by the effect of bamboo fillers in EPDM/rNBR has been broad to the addition of compatibilizer in EPDM/rNBR blend. The purpose of compatibilization was to increase the compatibility and homogeneity of EPDM/rNBR blends. Compatibilizer trans-polyoctylene rubber (TOR) were added into EPDM/rNBR filled bamboo fillers blends at different formulation and blend ratios respectively (2, 4, 6, 8, 10 phr). The cure characteristics, tensile and