UNIVERSITI PUTRA MALAYSIA

EFFECTS OF PARTICLE SIZE AND WEIGHT PERCENTAGE OF WASTE RUBBER CRUMBS ON THE PERFORMANCE OF COMPOUNDED TYRES

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By

ADNAN ABBAS ABDULNABI

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfillment of the Requirements for the Degree of Doctor of Philosophy

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DEDICATION

This thesis is dedicated to:

The sake of Allah, my Creator and my Master,

My great teacher and messenger, Mohammed (May Allah bless and grant him and his family),

My humble effort I dedicate to my sweet and loving

Wife (Intedar)

I am grateful to her support ....

To my daughter (Dalia) and son (Redah)

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Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfillment of the requirements for the Degree of Doctor of Philosophy

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By

ADNAN ABBAS ABDULNABI

February 2018

Chairman : Faieza Abdul Aziz, PhD
Faculty : Engineering

The problem of waste tyre disposal has become a seroius global issue as it has affected not only ecnomy, but also ecology of nations. As such, recycling provieded a powerful solution for the incresing of worn out tyres dumped into the landfills. At current, the recycled rubber produced still has vulcanized rubber structure and low blending ability with the virgin matrix composites. This behaviour limited the scope of engineering applications which could potentially involved recycled rubber material. The aim of this research was to study the effects of waste rubber modification such as size reduction on the manufacturing process of the tread of the passenger tyre. The study followed by construction of finite element code to predict the static and dynamic effects of the tyre material design and modification. Lastly, an empirical model was established to describe the effect of waste crumb rubber incorporation in tyre tread blend. In achieving the first objective Crumb Rubber (CR) modification was investigated, which was the size reduction method. A CR from ambient grinding of the ground tyre was used after it had been subjected to sieve analysis to produce six size categories of 40 µm, 150 µm, 180 µm, 250 µm, 425 µm and 600 µm. Each category was incorporated in different fractions 20 phr, 40 phr and 60 phr (10, 20, 30 wt.%) as a filler in a virgin styrene butadiene rubber (SBR) matrix to examine the effect of CR loading on the properties of the blends produced. The results revealed that the mechanical and curing properties of the tread blend had improved as the CR particle size reduced, with all of the mechanical and rheological

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properties reduction in a different manner when the CR content increased. The study revealed that the size reduction of CR could enhance the tensile strength up to 35%, and 17% for elongation at break for the finer size. To study the dynamic behavior of the tyre containing such modifyed CR, a finite element model based on Abaqus software was built as the second objective of the study. The model was used to study the effects of different particle size and content of CR on the traction, braking and slipping condition of the tyre. The parameters such as reaction force (rolling resistance), reaction moment, and pressure at footprint, stress, strain and strain energy were studied to figure out the complete behavior of the tyre at different tread properties. Finally, empirical models for tensile strength as a function of particle size and content of CR were built. Particle swarm optimization (PSO), and response surface method (RSM) based on design expert software were used. The results showed that both models have good accuracy in predicting the response, however the RSM model provided more accurate results. The statistical analysis confirmed that the effect of particle size was greater than the effect of content of CR on tensile strength of the tread.

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Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Doktor Falsafah

KESAN SAIZ ZARAH DAN PERATUSAN BERAT SISA SERBUK GETAH KE ATAS PRESTASI TAYAR BERKOMPAUN

Oleh

ADNAN ABBAS ABDULNABI

Februari 2018

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Kebelakangan ini, isu berkaitan pelupusan tayar sisa menjadi isu global yang semakin serius, ianya bukan sahaja menjejaskan ekonomi, bahkan ekologi sesebuah negara. Oleh yang demikian, kitar semula merupakan penyelesaian yang utama dalam menangani masalah pertambahan tayar buangan di tapak pelupusan. Pada masa kini, getah kitar semula yang dihasilkan masih mempunyai struktur getah tervulkan dan keupayaan penggabungan yang rendah dengan komposit matriks asli. Sifat ini menghadkan lagi bidang aplikasi kejuruteraan yang membabitkan penggunaan bahan getah kitar semula. Tujuan kajian ini adalah untuk mengkaji kesan pengubahsuaian sisa getah seperti pengurangan saiz pada proses pembuatan ke atas bunga tayar penumpang. Diikuti dengan penghasilan kod elemen tetap untuk menetukan kesan statik dan dinamik dalam reka bentuk dan pengubahsuaian bahan tayar. Dalam mencapai objektif pertama, pengubahsuaian serbuk getah (CR) telah disiasat, yang merupakan kaedah pengurangan saiz. CR dari pengisaran permukaan tayar selepas menjalani analisis ayak digunakan untuk menghasilkan enam kategori saiz 40, 150, 180, 250, 425, dan 600 µm. Kemudian, setiap kategori dicampurkan ke dalam pelbagai campuran dengan 20, 40 dan 60 phr (10, 20, 30 peratusan berat) sebagai pengisi dalam matriks getah asli stirena butadiena (SBR) untuk mengkaji kesan penambahan CR terhadap sifat-sifat campuran yang terhasil. Keputusan menunjukkan bahawa sifat mekanikal dan sifat pengawetan bagi campuran tayar bertambah baik apabila saiz zarah CR berkurang, dengan semua pengurangan sifat mekanikal dan reologi dengan cara yang berbeza apabila kandungan

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CR meningkat. Tambahan lagi, pengurangan saiz CR dapat meningkatkan kekuatan regangan sehingga 35% dan pemanjangan pada takat patah sebanyak 17% untuk saiz yang lebih halus. Model elemen tetap berdasarkan perisian Abaqus dibina untuk menyiasat kesan pengubahsuaian tayar bunga terhadap sifat dinamik tayar. Model ini digunakan untuk mengkaji kesan saiz zarah yang berbeza dan kandungan CR pada daya tarikan, cengkaman dan gelinciran tayar. Parameter yang dikaji adalah seperti daya tindak balas (rintangan putaran), momen tindak balas, tekanan cetakan, tekanan, tegangan dan daya tegangan untuk mengetahui sifat-sifat keseluruhan tayar pada komposisi yang berlainan. Akhirnya, model empirikal untuk kekuatan tegangan sebagai fungsi untuk saiz zarah dan kandungan CR dibina. Kumpulan zarah optimum (PSO) dan kaedah tindakbalas permukaan (RSM) berdasarkan perisian pakar reka bentuk telah digunakan. Keputusan menunjukkan bahawa kedua-dua model mempunyai ketepatan yang baik dalam meramal tindak balas, namun begitu model RSM menunjukkan keputusan yang kurang tepat. Analisis statistik mengesahkan kesan saiz zarah adalah lebih besar daripada kesan kandungan CR terhadap kekutan regangan tapak. Bagi kaedah eksperimen terdahulu yang berkos tinggi.

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I would like to dedicate this thesis to the memory of my parents who paved the path of knowledge upon their shoulders before I became who I am now. Priceless gratitude to my wife, Intedar for her great sacrifices, understanding and patience throughout the whole of our life together, which has made this study possible. Thanks to my lovely children who have also given a lot of moral support and encouragement for the whole duration of study in Malaysia.

Adnan Abbas Alshukr

This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfillment of the requirement for the Degree of Doctor of Philosophy. The members of the Supervisory Committee were as follows:

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Declaration by graduate student

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LIST OF ABBREVIATIONS

6PPD	Permanax
ACOH	Glacial Acetic Acid
ASTM	American Society For Testing And Materials
C_{ij}	Rivlin coefficient
D_i	Material Compressibility
CBS	N-Cyclhexyle-2-Benzothiazyl Sulphenamide
CR	Crumb Rubber
CRI	Cure Rate Index
CTP-100	Phthalimide
DOE	Design of Experiment
DR	Devulcanized Rubber
ELT	End of Life Tyre
EPDM	Ethylene Propylene Diene Monomer
ETRMA	European Tyre & Rubber Manufacturers Association
GTR	Ground Tyre Rubber
LLDPE	Linear Low-Density Polyethylene
MAE	Mean Absolute Error
MAPE	Mean Absolute Percentage Error
MH	Maximum Torque
ML	Minimum Torque,
PSO	Particle Swarm Optimization
R	The universal gas constant
RMSE	Root Mean Square Error
RSM	Response Surface Method
SBR	Styrene Butadiene Rubber
T	The thermodynamic temperature
TCA	Trichloroacetic Acid
TMQ	Flectol
Tol	Toluene

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LIST OF NOMENCLATURES

C_{ij}	Rivlin coefficient
D_i	Material Compressibility
G	Shear modulus
I	Principal strain Invariance
ts_2	Scorch Time
ts_{90}	Curing Time
υ_x	Tyre Rolling Longitudinal Velocity
υ_y	Tyre Cornering Velocity
V_e	Cross link density
W	Elastic Strain Energy Density
wt	Crumb Rubber Content
λ	Principal Stretch Ratios
σ	Stress
σt	Tensile Strength
σ_U	Uniaxial Nominal Stress
ω	Angular Velocity
ε	Strain

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CHAPTER 1

INTRODUCTION

1.1 Introduction

In the new global ecology, the problem of waste tyres has become a central issue. Millions of worn out tyres are being thrown in the tyre graveyards every year all over the world. The worldwide number of waste tyres in storage has exceeded three billion [1]. According to a previous estimation, around 800 million of scrap tyres have been disposed of all around the globe. This amount is expected to increase by approximately two percent each year [2]. The waste tyre stockpiles growing has been attributed to the tremendous increased rate of tyers production, which had reached 1.72 billion units at the end of 2015 according to the statistics from the global industry analysis [3]. Tyers are highly engineered and have three-dimensional chemical network constructed from many dissimilar materials. Rubber (natural or synthetic) represents the matrix of the tyres composite structure, which makes up the biggest component used in tyres manufacturing as shown in Table 1.1.

Table 1.1 : Major composition of material used in tier manufacturing [3]

Ingredient	Car/passenger	Truck	Other
Rubber	47%	45%	47%
Carbon black	21.5%	22%	22%
Metal/ steel	16.5%	25%	12%
Textile	5.5%	0%	10%
Zinc Oxide	1%	2%	2%
Sulphr	1%	1%	1%
Additives	7.5	5%	6%

The rubber stabilizer materials, such as antioxidants, antiozonants and other additives present in the tyre manufacturing process lead to producing high cross-linked chemical structures rubber that is required to overcome the different operating conditions. These structures cause the waste tyre to be a non-biodegradable and non-environmentally friendly material [4]. The landfilling and mono filling were the common earlier ways of tyre disposal over the world, for example the United States and Europe reclaimed just about one-fifth of rubber hydrocarbon used at the end of the 1950s and most of the

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end of life tyers (ELT) were discarded in landfills [5]. This method of tyre disposal is undesirable because of the following reasons:

- The probability of contaminating the surrounding soil due to tyre additives leaching
- Represents negative added value because of implicit cost of transportation of tyres to the landfill sites and maintenance of the landfills to satisfy the environmental requirement.
- Tyre shape and permeability produce a long time water holding which provides sites for rodents, snakes and insects breeding [6].
- The amount of rubber and other metal contents present in the ELT makes the landfilling process represents a disposal of valuable materials.
- The landfilled ELT tyres possess fire threat danger which is difficult to be extinguished [7].

Therefore, recycling and recovering of ELT is the best economical and ecological solution to the tyre disposal problem. Recently, legislation has driven the tyre section toward recycling and recovery. The issue of ELT treatment has received considerable critical attention during the last decade. The rate of treatment has increased rapidly as the environmental awareness has increased. A series of law and regulations have been implemented from 48 states to govern the collection, handling and disposal of the ELT [8]. As so, the rate of ELT treatment had increased from 10 percent to 80 percent during the period from 1990-2012 for the United State of America, while Europe had reported being the highest in the world at the end of 2012, by transferring around 95 percent of ELT to be recycled and recovered into materials as shown in Figure 1.1[9].

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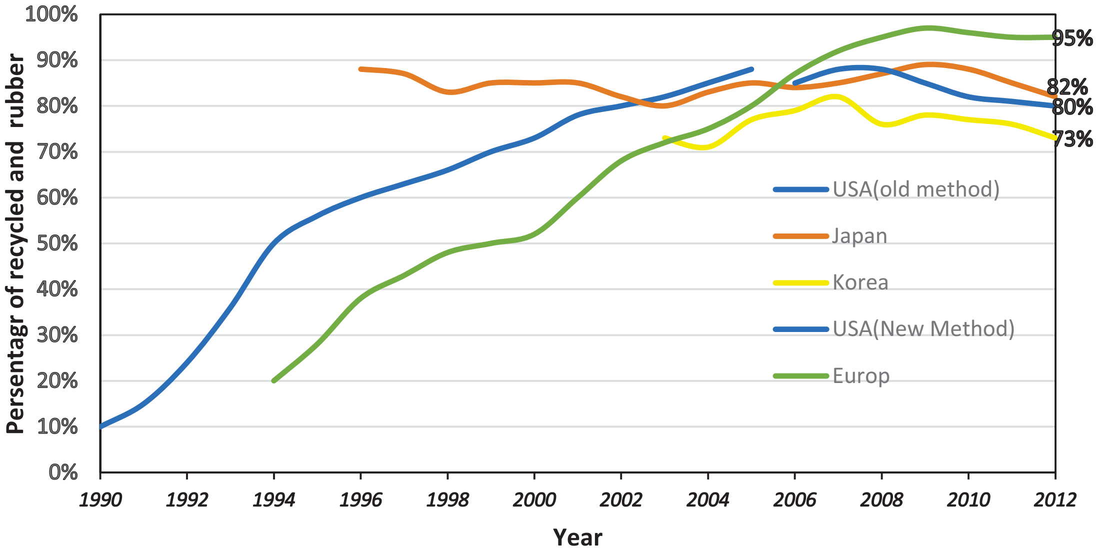

Figure 1.1 : The percentage of recycled and recovered tyres relative to the ELT [9]

According to the report published by the European Tyre & Rubber Manufacturers Association (ETRMA) in EU member states, Norway, Switzerland and Turkey [10], around 3.868 million tons of used tyres were generated in 2015. After sending 678 thousand tons for reuse, export and retreading, 3.190 million tons of ELT were sent to the recovery and recycling. The rate of recovery estimated was 92 percent, which represents one of the highest rates in recovery and recycling of ELT in the world.

It is evident from this report that, there is a global concern and urgency to minimize the amount of landfill tyres to its lowest and manageable level and moved towards a zero-waste scenario. This level had already been reached in most of the Europe states, but some efforts remained to be done through the development of the current recycling methods and find new techniques to utilize the produced recycled rubber in a different application. Although, around 50 percent of waste rubber are recycled and converted to ground rubber to use as raw materials for many applications, like civil engineering projects, new tyre production and many other rubber base applications, there is still, according to the ETRMA report [10], about 35 percent of waste tyres are used as fuel. This is harmful to the environment and this material needs to be converted for reuse or recycle, instead of using it as fuel in support of sustainability. This effort requires continuous research and developments to innovate new devulcanization and reclaiming methods in order to strategically manage and incorporate the use of crumb rubber (CR) in current applications as well as finding new engineering applications.

High-energy irradiation offers unique solutions to the problem of recycling due to its ability to induce crosslinking or scission of a wide range of material without introducing any chemical initiators. This method can possess a significant economical and ecological advantage as compared to the conventional chemical, thermal and mechanical methods which may cause many drawbacks like, noxious fumes emission, by-products peroxide degradation and high energy consumption. The three main possibilities for the use of radiation in this application are:

- Enhancing the mechanical properties and performance of recovered materials or material blends, principally through crosslinking, or through surface modification of different phases being combined.
- Treatment causing or enhancing the decomposition of polymers, particularly through chain scission, leading to the recovery of either low molecular weight mixtures, or powders, for use as chemical feedstock or additives.
- Production of advanced polymeric materials designed for environmental compatibility.

Therefore, irradiation represents an attractive solution to the problem of recycling of waste rubber in polymer composite field.

The use of numerical analysis techniques as the finite element method (FEM) has become quite a convenient and powerful tool in mechanical engineering, particularly for product design and development. Also in tyre researches, the determination of the main trends and designs is performed by means of numerical simulations and comparative evaluations. The most crucial aspect of a tyre simulation is the realistic description of the processes in the contact interface between the tyre’s tread and the road surface. The frictional behaviour of this interface controls the performance of the tyre not only locally, but also globally. As such, the FEM can play a major role in tyre structural and material design process through its ability to simulate the static and dynamic performance of the tyre in different working condition.

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1.2 Problem Statement:

Every year the amount of worn out tyres are increasing over the world, however the development of the recycling process does not keep up at the same rate. Most of the recycling factories are facing real commercial problems, due to lack and limit of its products applications.

Crumb rubber is the main final product of recycling rubber factories. The main function that decides the ability of any filler to be incorporated for certain application is the active surface area[11]. This area represents the trend of filler to create crosslink with matrix materials. In general, crumb rubber has low ability to build a suitable bond with any other material, because of the strong sulphidic and carbon molecular bonds. Several surface treatments have been developed in targeting to reactivate crumb rubber surface area. The chemical activation process comprises of grafting, swelling, halogenation and addition of polymer or curing system, are examples of using chemical reaction in surface modification of CR. In addition to the chemical process, there are other numerous methods based on their physical activation such as microbial treatment and superficial devulcanization [12]. Nevertheless, all of the above methods are costly and by incorporating the process will affect the recycling factories economically.

One of the most important usages of CR is being a filler as a replacement of expensive virgin rubber matrix, this phenomenon is used in tyre tread blend as a part of manufacturing of passenger and truck tyres. The amount of CR used in this blend does not exceed 10 percent of overall weighted percentage according to the manufacturing documents of Dunlop [13]. Cheap and active methods of CR modification may lead to an increase in the amount of costless CR against high-cost virgin rubber in tread tyre blend and in other virgin polymer matrix. In addition, the allowable limit of CR particle size and content in tread compound is an important issue. However, there is lack of literature on the effect of using the particle size and content that can be incorporated in the tread of the passenger tyre.

Changing the amount of any material in tyre blends recipes are costly and time-consuming procedure. This process passing through series of activities and steps comprise internal and external tests equipment to predict the influence of this change or design in the final performance of the tyre. The other way to avoid this problem is by importing new technology from one of the tyre knowhow companies,

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which is very expensive and almost not flexible solution. The finite element code may provide a feasible improvement to the process of testing the new material design, if it is accompanied with basic laboratory mechanical test for the new design. The most crucial aspect of tyre simulation is the realistic description of the process in the contact interface between the tyre tread and road surface [14]. The dynamic behaviours of the tyres represent further challenging because it depends on many factors, such as tyre type (radial or bias-ply), tyre structure (geometry, reinforcement and tread pattern), loading condition (inflation pressure and wheel load), tyre rolling condition (traction, free rolling , braking and cornering). As such, a suitable and accurate finite element model would provide significant benefits to show the comparative interface under various tyre loading conditions [15].

In summary, the problem statements of this work can be summarized as follow:

1. The waste tyres represent a serious ecological problem and negative added cost which needs extensive support to address.
2. The existence of strong sulphidic and carbon molecular bonds causes the limit applications of crumb rubber.
3. Limited amount of cheap crumb rubber (10% of tread layer weight) is used to replace the expensive virgin rubber in tyres manufacturing application.
4. The reclaiming and devulcanizing methods needed to recycle the waste rubber are costly methods, as such there is a need to develop the current method by introducing new cost effective methods.
5. There is a need to draw a complete perception to the effect of using the particle size and content that can be used in the tread of the passenger tyre.
6. The assessment process for a new design of a tire material by using the traditional experimental methods is difficult and expensive efforts.

1.3 Objectives

The primary aim of this study is to determine experimentally the effect of crumb rubber modification on the rubber blend through one of its important application “tyre tread” as a contribution to increase its content in the tread recipe to achieve goals of reducing consumption of natural resources and supporting waste tyres recovery process, which represents eco-friendly activity. The particular objectives of this work are:

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1. To investigate the effect of CR modification such as particle size and content of waste rubber crumb on the passenger tyre tread composite material.
2. To analyze the dynamic performance of a passenger tyre tread compounded with waste rubber crumb.
3. To develop particle swarm optimization and response surface method models for the tensile strength of tyre tread containing specific particle size and weight of waste rubber crumb.

1.4 Scope of Study

This study focuses on determining of new method of modification of CR. This process, which represents the first objective, may lead to an increase in the CR loading amount in a compound recipe without affecting the final properties of the composite. The CR was incorporated in the recipe of passenger tyre tread, which has been chosen as the application to find the efficiency of the process. In this investigation, six sizs (40, 150, 180, 250, 425 and 600) µm of CR particle size was incorporated with CR loading of 10, 20 and 30 weight percentage of the total blend recipe for each category. The mechanical and rheological study for each particle size and content was performed to find the trend of each parameter. For the second objective, a finite element model of passenger tyre based on Abaqus package was built to predict the static and dynamic performance of the tyre containing the modified tread material. The material properties of the model are based on data extracted in the laboratory to analyze its performance when it becomes part of a real tyre structure. Finally, a numerical model for tensile strength of the tread compound was built. The model is created based on the data extracted from the first object by using two different methods, particle swarm algorithm and response surface method.

1.5 Thesis Outline

This research consists of five chapters:

Chapter 1: This chapter presents a brief background on the field of waste tyre problems and recycling challenges. The chapter describes the importance of finding new reclaiming and surface activation methods for end of life tyres to ensure sustainability for the future. In addition, the research problem was highlighted and the objective of research was defined within the boundary of this work.

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Chapter 2: This chapter contains reviews of the available literature on the methods used to recycle waste tyres with emphasize on the recovery of rubber material and a brief discussion of methods used to activate and modify the CR with detailed discussion in irradiation and size reduction methods, which are the methods used in this study. This chapter also provides a literature review on the available researches in the field of tyre analysis and the main constitutive equations used to describe the hyperplastic materials.

Chapter 3: In this chapter, an elaborate discussion regarding the materials specifications, characterizations, mechanical and rheological assessment details will be covered, in addition to the process and condition of blends preparation method. This chapter also contains a detailed description of the special techniques that were carried out to build the finite element model of the passenger tyre. This part of methodology provides the Abaqus code details and boundary conditions for each step of the solution. Finally, the process of creating an empirical model for the tensile strength of the tread was described. The model was built by using the numerical algorithm based method, which is particle swarm optimization method and mathematical and statistical based method namely surface response method.

Chapter 4: The chapter is dedicated to discuss further the results and findings that emerged from the study with appropriate cause and effect for each particular outcome.

Chapter 5: A conclusion on the findings of the research will be drawn in this chapter. Finally, the recommendations for future research will be suggested.

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