EFFECT OF NATURAL FILLER ON THE CHARACTERIZATION OF NATURAL RUBBER

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EFFECT OF NATURAL FILLER ON THE CHARACTERIZATION OF NATURAL RUBBER

SANTINI RAJ A/P KAMARAH

A thesis submitted to the Faculty of Chemical and Natural Resources Engineering In Partial Fulfillment of the requirements for the award of the Degree in Bachelor of Chemical Engineering

Faculty of Chemical & Natural Resources Engineering
Universiti Malaysia Pahang

NOVEMBER 2010

I declare that this thesis entitled "Effect of Natural Filler on the Characterization of Natural Rubber" is the result of my own research except as cited in references. The thesis has not been accepted for any degree and is not concurrently submitted in candidature of any other degree."

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Name : **SANTINI RAJ A/P KAMARAH**
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To My Beloved Father and Mother

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ABSTRACT

In this paper the effect of natural filler on the characterization of natural rubber will be investigated. Usage of natural rubber only to produce rubber products becomes disadvantages in economical and environmental aspect. It will be more efficient if the rubber is reinforced with filler as its additive without affecting its properties. Moreover, the increase interest in using natural fillers as reinforcement in natural rubber to substitute the conventional fillers has become one of the main concerns in nowadays. For this study natural rubber that been used is Standard Malaysian Rubber (SMR) while coconut fiber is the natural filler that being use to reinforced the SMR. The composite material using natural rubber and coconut filler are being done in two methods; first method is by mixing by using two roll mills to the natural rubber without any additional of fillers. The second method is to mix homogeneously the natural rubber with different loading (0-50phr) of coconut fiber. Later chemical substances such as sulphur, softener, accelerator and etc were added using rolling process according to the formulation. The cured rubber is molded using hot press and sample is cut manually from the molded rubber for the testings. As the filler loading increases, the time consumed to blend the SMR and filler are getting long because the volume of coconut fiber is increasing. The mixing or rubber and other additive are also getting tougher as the filler loading increasing. The tensile strength is decreasing as the filler loading increases. Besides that, extension at break and time at break is decreasing when the filler loading is increasing. For swelling test, kerosene diffuse more than diesel in the reinforced rubber but decreasing as the filler loading increases. It is concluded that the new composite material using natural rubber and coconut filler giving less strength than using conventional filler. In conjunction of that, using of natural filler as reinforced for rubber is best for the products that need less strength.

ABSTRAK

Kesan daripada pengisi semulajadi terhadap perwatakan getah asli dikaji dalam penyelidikan ini. Penggunaan getah asli sahaja dalam produk berunsurkan getah boleh merugikan dari segi ekonomi dan juga alam sekitar. Dengan ini, adalah efisien jika getah asli ditambahkan dengan pengisi sebagai bahan tambahan dalam pemprosesan tanpa mengusik sifat yang terdapat dalam getah asli. Selain itu, banyak kajian dilakukan terhadap pengisi semulajadi daripada pengisi lazim. Dalam kajian ini, getah asli yang digunakan adalah getah standard Malaysia (SMR) dan sabut kelapa sebagai pengisi semulajadi yang boleh menjadi bahan penguat kepada getah asli. Dua kaedah dilakukan iaitu pertamanya kajian dengan getah asli yang tidak ditambahkan pengisi dengan menggunakan mesin ‘Two Roll Mill’. Keduanya pula ditambah dengan sabut kelapa yang mempunyai variasi pemuatan dari 0-50phr. Kemudian, bahan kimia seperti sulfur, pelembut, pemecut dan lain-lain akan ditambah kepada komposit tadi mengikut formula yang telah ditentukan. Getah yang telah dirawat dibentuk dengan menggunakan mesin ‘Hot Press Molding’ dan sampel potong mengikut bentuk yang diperlukan untuk analisa. Semakin meningkat pemuatan pengisi, masa yang diambil untuk proses penambahan pengisi menjadi lebih panjang. Proses penggaulan getah dan bahan penambah juga menjadi semakin sukar. Kekuatan tegangan berkurang dengan penambahan pengisi. Selain itu, pemanjangan dan masa di waktu putus berkurang dengan penambahan pengisi. Bagi analisa pembengkakkan, minyak tanah meresap masuk ke dalam getah lebih banyak dari diesel tetapi berkurang dengan penambahan pengisi. Sebagai kesimpulan, getah asli yang ditambahkan dengan pengisi semulajadi memberi kekuatan yang rendah berbanding dengan pengisi lazim. Tambahan pula, getah asli yang ditambahkan dengan pengisi semulajadi adalah lebih padan digunakan untuk produk getah yang memerlukan kekuatan yang rendah.

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LIST OF ABBREVIATIONS

SMR	-	Standard Malaysian Rubber
EFB	-	Empty Fruit Bunch
RM	-	Ringgit Malaysia
NR	-	Natural Rubber
RSS	-	Ribbed smokeed sheet
SIR	-	Standard Indonesia Rubber
SR	-	Synthetic Rubber
cm	-	Centimeter
mm	-	Milimeter
µm	-	Micro meter
MPa	-	Mega Pascal
mm min^{-1}	-	Milimeter per minute
TS	-	Tensile Strength
E	-	Tensile Modulus
e	-	Elongaton at break
ASTM	-	American Society for Testing and Materials
Corp	-	Corporation
Phr	-	Part per Hundred
CaCO3	-	Calcium Carbonate
ZnO	-	Zinc Oxide
Zn	-	Zinc
IUPAC	-	International Union of Pure and Applied Chemistry
$C_{18}H_{36}O_2$,	-	Stearic Acid
6PPD	-	N-(1,3-Dimethylbutyl)-N'-Phenyl-P-Phenylenediamine
CBS	-	N-Cyclohexyl-2-Benzothiazole Sulfenamide
S	-	Sulphur
e.g	-	Example given

µ - Micro

kN - Kilo Newton

ml - Mililiter

hrs - Hours

L_f - Fracture Length

L_0 - Original Length

i.e - In Example

CHAPTER 1

INTRODUCTION

1.1 Background of Study

This study is based on the Effect of Natural Filler on the Characterization of Natural Rubber. Natural rubber is from the isoprene monomer (2-methyl-1,3-butadiene). It is an example of elastomeric type polymer, where the polymer has the ability to return to its original shape after being stretched or deformed. Besides that, natural rubber is an addition polymer that is in a form of milky white fluid known as latex from the rubber tree. The latex is obtained from the rubber tree through tapping a cut in the bark, precipitated with acid, then washed and dried. The natural rubber has low tensile strength, low elasticity, can't withstand high temperature, absorbs a large amount of water, low abrasion resistance, and etc. There are few grades for natural rubber such as Standard Malaysian Rubber (SMR), Standard Indonesia Rubber (SIR) where SMR can be sub graded from SMR 5, 10, 20 and 50.

Though, natural rubber can be reinforced by adding fillers. Fillers are solid substance that is added to the rubber to increase the strength usually increasing hardness and durability. Moreover it is to reduce the cost of rubber compounding. The uses of the fillers are as old as the uses of rubber itself. Fillers can be found in two types. They are natural fillers and also synthetic fillers. Carbon black, precipitated silica, barite, talc and calcium carbonate are the examples of synthetic fillers meanwhile rice husk, coconut fibers, and empty fruit bunch (EFB) are the examples of natural fillers. Natural fillers are biodegradable, environment friendly and easy to get because it is in abundant quantity. In addition, natural fillers are cheap to purchase and can reduce the domestic waste. So, it will be credit to choose natural filler as the additive. I n this study, coconut fiber has been chosen as the filler for the natural rubber. It mainly contains of cellulose, hemicelluloses, lignin, and other vital substances. Some properties of coconut fiber are low density, high degree in retaining water, tough, stiff and etc.

1.2 Problem Statements

Natural Rubber normally uses in industry that producing automobile tires, gaskets, hoses, personal protective equipments and etc. The demands for rubber based products are increasing by time. Meanwhile the current price of natural rubber based on Malaysian Rubber Board is about RM12 per kg and is increasing by day. So, in economical view, it is not possible to produce rubber products by just using natural rubber only. In this point, some materials are needed which could be mixed with natural rubber to produce same or better quality products up to the consumers' expectation. By that, it is found that fillers can be the additive agent and also functioning to improve the strength of natural rubber. In conjunction, natural fillers can be chosen to be reinforced with the natural rubber as they can reduce wastes from landfills. For that, coconut fiber is one of the natural fiber that are being waste daily. So, by finding possible applications for coconut fibers that would otherwise become waste, it can be used in reinforcing the natural fiber and increase the mechanical properties of the rubber itself. Furthermore, it may be a cheaper alternative in the future to process the natural rubber.

1.3 Objectives

The objectives of this study are as follow:-

i. To study the effect of filler loading to the natural rubber.
ii. To compare the strength of reinforced natural rubber with the original natural rubber.
iii. To study the effect of mixing time to filler loading.

1.4 Scope of Study

The scope of project for this study Effect of Natural Filler on the Characterization of Natural Rubber is limited to certain limitation. In this case, the limitation of the study is limited on natural fillers that reinforce the natural rubber. More specifically, the natural filler that been chosen for this study is coconut fiber and for natural rubber, Standard Malaysian Rubber-10 (SMR10) has been chosen. Besides that, effect of mixing and molecular structure of reinforced rubber also being discussed in this study. The chemistry of rubber compounding will be explained in detail. This study also limited to have testing such as tensile test and swelling test.

1.5 Significance of Study

The study on this topic can be benefit for certain circumstances. First of all, it is an environmental friendly project where the wastes produce by coconut can be reduced and turning it to a wealth product. Besides that, it helps to reduce the amount of natural rubber used in the production as it will be a disadvantage in economical wise and bring lose to the manufacturers. This study is possible to commercialize and be a cheaper alternative in the future of rubber technology as there were no many researches on this specific topic. Moreover, it is not to be tested yet.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

In this chapter, basically it describes more about the Effect of Natural Filler on the Characterization of Natural Rubber that has been done by the other researchers from industries and institutes. Therefore in this part, first of all, the discussion will be focused on natural rubbers and natural fillers. While in second part, it will be on the chemistry in mixing of compound and flow of behaviour. Later, the tests done to evaluate mechanical properties of reinforced natural rubber also included in this chapter such as tensile test and swelling test.

2.2 Rubber

2.2.1 History and Development

Natural rubber also has been called ‘the supreme agricultural colonist of all times’. It is originally indigenous from the Amazon Valley forest, but has been cultivated principally in Southeast Asia, especially for countries like Malaysia and Indonesia. Where more than fourteen million acres of land have been cleared and planted with rubber trees (Yam, K.P., 2007).

Priestley, the English chemist, gave the name ‘rubber’ to the raw material in 1770, when he found it would ‘rub off’ pencil marks. In early 1800’s, a Scotsman, Macintosh, patented the first raincoat, consisting of a layer of rubber sandwiched between two thicknesses of cloths. A London, coachman Thomas Hancock, then cut rubber into long strips, and invented the ‘rubber band’ in 1823 (Yam, K.P., 2007).

In 1839, the tremendous discovery of vulcanization occurred, which ultimately made possible the multibillion dollar industry of today. The combination of sulphur dust and heat added to raw rubber resulted in remarkable resistance of the ‘vulcanized’ rubber to extreme temperature changes has found by Hancock and Charles Goodyear (Yam, K.P., 2007).

2.2.2 Natural Rubber

Natural Rubber can be described as a material which is highly elastic where it can be stretched without breaking and will return quickly to its original length on releasing the stretching force **(**Yam, K.P., 2007**).** Natural rubber is poly-cis-1,4-

isoprene, and is highly stereo regular. Isoprene (2-methyl-1,4-butadiene) has the structure as Figure 2.1:

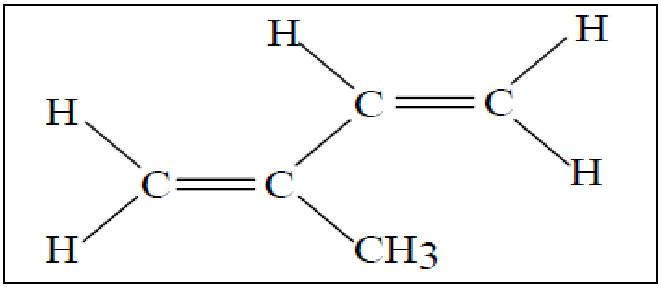


Figure 2.1: Isoprene

and the polymer is as in Figure 2.2:

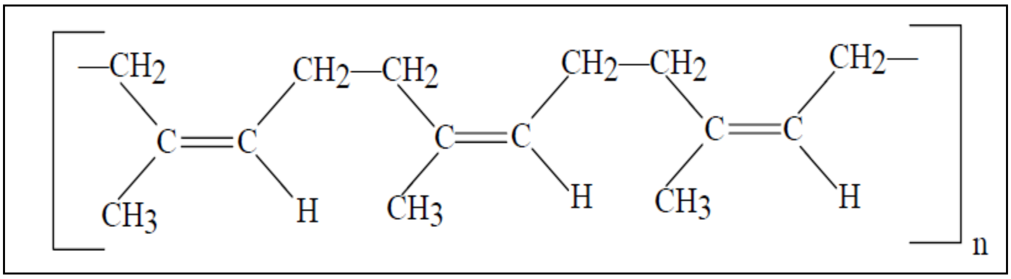
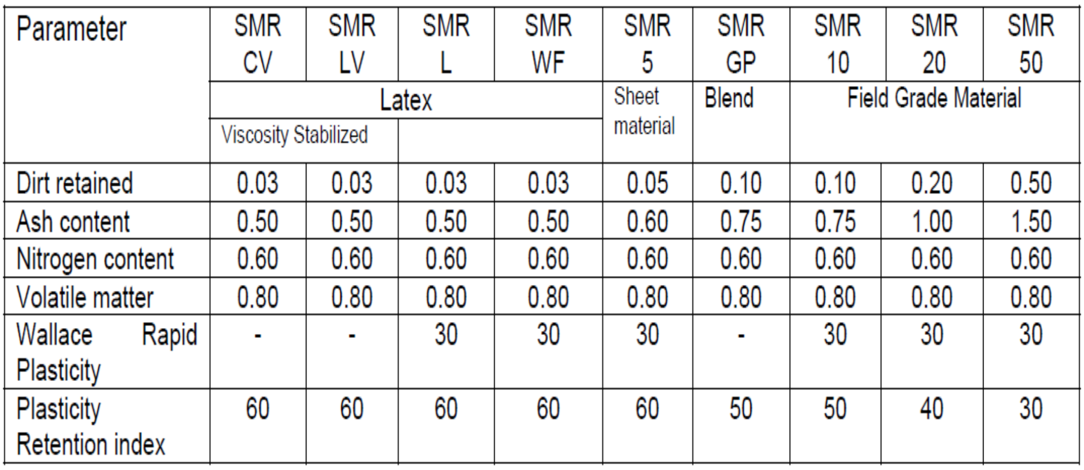


Figure 2.2: Polyisoprene

Natural rubber hardens below 0^0C and softens and weakens above 80^0C, losing its strength and becoming tacky. In between these temperatures it can flow under stress and permanent deformation occurs under prolonged strain. These undesirable properties are reduced by vulcanization, in which the reactivity the double bonds impart to the molecule is utilized to make it react with added material to form cross links between the chains. The cross linking increases the useful temperature range of the rubber and hardens the rubber so that it becomes much stronger and does not creep but returns to its original shape on release of stress. Its surface properties are improved and its solubility decreased. Sulphur is still the most important vulcanizing compound for natural rubber, but not the only one.

Although the chemical building block of the polymer is always the same, i.e. polyisoprene. Natural rubber (NR) is available in many grades related to its ‘dirt’ content (comes from tree) and precise method of production. Popular grades are ribbed smoked sheet (RSS) and technically specified rubber such as SMR (Standard Malaysian Rubber) and SIR (Standard Indonesia Rubber). All of these are sub graded by dirt content. Grades are RSS1 to RSS5 and SMR 5, 10, 20 and 50; the lower number, the cleaner of the grade, and therefore the more expensive. Natural rubber and synthetic elastomers are commercially available in different viscosity grades. Lower viscosity elastomers are often chosen to ensure easy flowing compounds in the manufacture of molded goods. Higher viscosity grades are often used in extrusion grades because they provide high green-strength, even when highly extended with fillers and oils. However, as has been stressed earlier, the primary consideration in choosing a type and grade of elastomer is that it should provide the properties required in the end application (Johnson, P.S., 2001).

Table 2.1: Standard Malaysian Rubber Specification (Hofmann, W., 1989).



Parameter	SMR CV	SMR LV	SMR L	SMR WF	SMR 5	SMR GP	SMR 10	SMR 20	SMR 50
	Latex				Sheet material	Blend	Field Grade Material		
	Viscosity Stabilized								
Dirt retained	0.03	0.03	0.03	0.03	0.05	0.10	0.10	0.20	0.50
Ash content	0.50	0.50	0.50	0.50	0.60	0.75	0.75	1.00	1.50
Nitrogen content	0.60	0.60	0.60	0.60	0.60	0.60	0.60	0.60	0.60
Volatile matter	0.80	0.80	0.80	0.80	0.80	0.80	0.80	0.80	0.80
Wallace Rapid Plasticity	-	-	30	30	30	-	30	30	30
Plasticity Retention index	60	60	60	60	60	50	50	40	30

Properties of vulcanized product made from natural rubber are as follows:
(Ciesielski, A., 1999)

- High mechanical strength and can be compounded to have excellent elasticity.
- Good abrasion resistance, low relative cost, significant choice for slurry pump liner and impellers as well as for tank lining.
- Good dynamic mechanical properties used in tires, rubber springs and vibration mounts.
- Electrical insulation is very good.
- Dilute mineral acid (although not oxidizing acids such as nitric) and dilute base resistance is good.

2.2.3 Application of Natural Rubber

Natural rubber is a very versatile raw material, had a widely application use on industry material and domestic. Natural rubber can be directly used for making adhesive tapes, rubber solutions or art gum but most is vulcanized. With the increasing and improvement of Synthetic Rubber (SR) grades, natural rubber has been slowly replaced in many applications which require heat and swelling resistance. But natural rubber is preferably used in large tyres due to its poor conductor of heat and heat build-up, always been of great important for producing truck tyres. With the introduction of passenger radial tyres and heavy radial truck tyres, the use of natural rubber has been increase. The excellent flexibility of natural rubber is suitable for sidewall compound and its low heat build-up results in cooler running tyres (Hofmann, W., 1989). Natural rubber also has important application in the production of thin walled such as balloons, surgical gloves or sanitary rubber product due to its strain crystallization and self-reinforcing properties.

Figure 2.3: Applications of Natural Rubber

Figure 2.4: Gloves

2.3 Fillers

2.3.1 Fibers

Fibers are class of hair-like materials that are in discrete elongated pieces, similar to pieces of thread (Muhammad Jannah, J., 2008). They can be spun into filaments, thread or rope. They can be used as a component of composite materials. Fiber can be classified into two main groups, which are man-made fiber and natural fiber. In general, natural fibers can be subdivided as to their origin such as plants, animals, or minerals; while man-made fibers can be subdivided to synthetic and natural polymers.

The first fibers used by man were natural fibers such as cotton, wool, silk, flax, hemp and sisal. The first man-made fiber was probably glass **(**Muhammad Jannah, J., 2008**).** Both natural and synthetic fibers (commonly known as man-made fibers) are now available and always being used as fillers in making a good properties of composites. The major fibers used till now can be classified into the groups given in Figure 2.4.

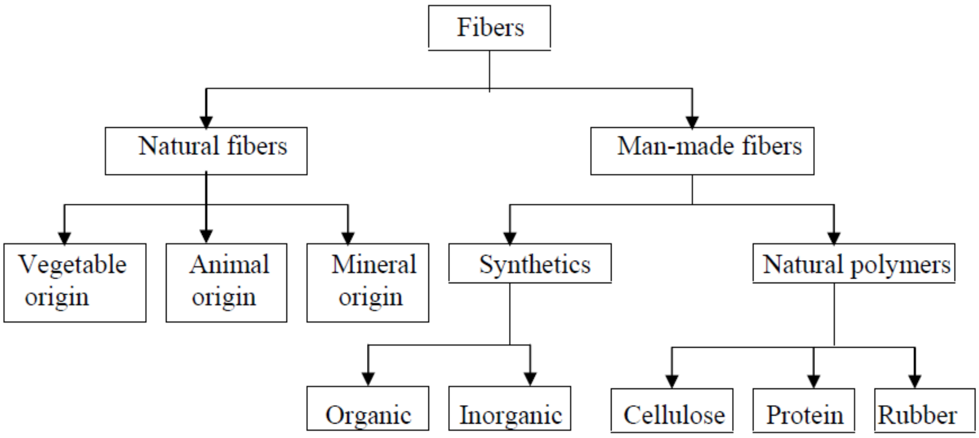


Figure 2.5: Classification of Fibers (Muhammad Jannah, J., 2008).

2.3.1.1 Natural Fiber

Natural fibers in the strict meaning of the word are produced through agriculture. Natural fibers are subdivided based on their origins, for example vegetable/plants, animals, or minerals. Vegetable or plant fibers include bast or stem fibers, leaf or hard fibers, seed, fruit, wood, cereal straw and other grass fibers **(**Muhammad Jannah, 2008). According to Muhammad Jannah, J., 2008, plants can stand up because of cellulose and lignin. Most plant fibers, except for cotton, are composed of cellulose, hemicelluloses, lignin, waxes and some water-soluble compounds, where cellulose, hemicelluloses and lignin are the major constituents.

Muhammad Jannah, J., 2008, summarized that the most important factor controlling the different types of natural fibers is their species because the properties of fibers are different between different species. In addition, the properties of fibers within a species vary depending on area of growth, climate and age of the plant. Lastly, the properties of natural fibers vary greatly depending on their processing method used to break down to the fiber level. Mineral fibers are naturally occurring fiber or slightly modified fiber procured from minerals. Mineral fibers such as asbestos fibers had been used historically for insulating houses. However, since January 1997, to provide protection of workers and consumers, the manufacture and transformation of asbestos fibers became forbidden (Bilba et al., 2007). Figure 2.5 shows the classification of natural fibers while Figure 2.7 shows the classification of vegetables or plant fibers.

Natural fiber provides sufficient mechanical properties, in particular stiffness and strength, at acceptably low price levels. Considering the ecological aspects of material selection, replacing synthetic fibers by natural ones is only a first step. Restricting the emission of green house effect causing gases such as CO2 into the atmosphere and an increasing awareness of the finiteness of fossil energy resources are leading to developing new materials that are entirely based on renewable resources (Tudu, P., 2009).

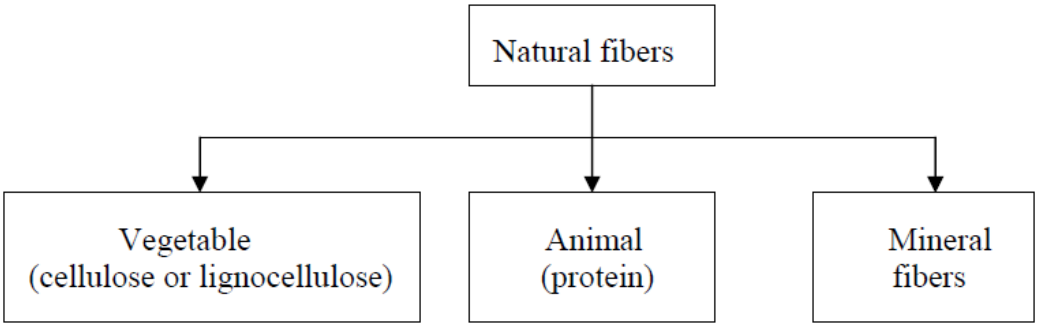


Figure 2.6: Classification of Natural Fibers (Muhammad Jannah, J., 2008**).**

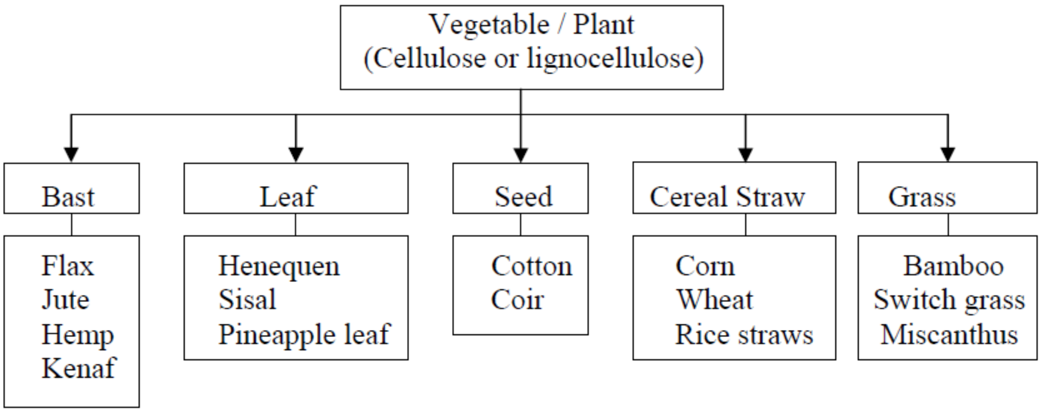


Figure 2.7: Classification of Vegetable or Plant Fibers (Muhammad Jannah, J., 2008**).**

2.3.1.2 Advantages of Natural Fibers

The advantages of natural fibers over synthetic or man-made fibers such as glass are their relatively high stiffness, a desirable property in composites, low density, recyclable, biodegradable, renewable raw materials, and their relatively low cost. Besides, natural fibers are expected to give less health problems for the people producing the composites. Natural fibers do not cause skin irritations and they are not suspected of causing lung cancer**.** The disadvantages are their relatively high moisture sensitivity and their relatively high variability of diameter and length. The abundance of natural fibers combined with the ease of their process ability is an attractive feature, which makes it a covetable substitute for synthetic fibers that are potentially toxic (Muhammad Jannah, J., 2008).

2.3.1.3 Coconut Fiber

Coir is processed from the husk of the fruit from the coconut tree (*Cocos nucifera*), which is a tall palm tree native to East India. Coconut fiber is one of the most important natural fillers produced in tropical countries like Malaysia, Indonesia, Thailand, and Sri Lanka (Sapuan, S.M., 2003).

The coconut husk is that 5-10 cm thick fibrous covering of the coconut fruit which envelops the hard shell structure of 3.5 mm thickness. The external appearance of the husk varies from decidedly dull brown when fully ripe to bright green when immature. There are other varieties whose husks are golden yellow or yellow brown. The husk is full of long, coarse fibers, all running in one direction. The fibers are embedded in a matrix of material called coir dust. Since husks are porous, they absorb or retain water (Tejano, E.A., 1985).

Many works have been devoted to use of other natural fillers in composites in the recent past and coconut filler is a potential candidate for the development of new composites because of their high strength and modulus properties. Composites of high strength coconut filler can be used in the broad range of applications as, building materials, marine cordage, fishnets, furniture, and other household appliances (Sapuan, S.M., 2003).

Table 2.2: Composition of Coconut Filler

CONSTITUENT	PERCENT
Moisture	15.0
Lignin	43.0
Ash	8.26
Alkalinity of ash (as K_2O)	37.5

In the extraction of coir fiber from the coconut husk and in the production of finished materials from the extracted fiber, a large amount of coir dust is produced. The coir dust is about 70% of the weight of the coconut husk. It is described as that brown, spongy particle of low weight which falls out when the fiber is shredded from the husk. The composition of coir dust is given in Table 2.3.

Table 2.3: Composition of Coir Dust

	PERCENT (DRY BASIS)	
CONSTITUENT	a	b
Moisture	15. 38	20. 0
Ash	6. 19	
Cellulose	24 .25	
Pentosan	27. 31	10. 4
Furfural	17. 40	
Lignin	54. 78	33.3
N		0.3
CaO		0.4
P_2O_5		0.5
K_2O		0.9

2.4 Mixing of Rubber Compounds

Mixing is the most critical component of rubber processing. Mixing requires deciding for a given formulation what equipment to use, and the times, speeds, pressures, temperatures and procedures that are required to blend those ingredients into an adequate mixed compound. This also requires consideration of costs.

The four main components of the mixing process are incorporation, dispersion, distribution, and plasticization or viscosity reduction.

i) Incorporation, sometimes called wetting, is the first stage in mixing, during which the previously separate ingredients form a coherent mass.
ii) Dispersion is the process during which filler agglomerates are fractured and reduced to their ultimate size.
iii) Distribution is simple homogenization, during which the various ingredients are randomly distributed throughout the mass of the mix.
iv) During Plasticization, the mix reaches its final viscosity as plasticizers effectively internally lubricate the mix.

These four processes are not entirely distinct. Distributive mixing, take place throughout the mixing cycle. However, incorporation predominates in the early stages, dispersion in the middle, and plasticization towards the end (Johnson, P.S., 2001).

2.4.1 Incorporation

Incorporation is the preliminary step in mixing, in which the initially separate ingredients from coherent mass. The step has two parallel mechanisms. In the first, as the elastomer undergoes deformation, it provides an increased surface area for accepting fillers agglomerates and then seal them inside. This can be readily visualized in mill mixing, where cutting and blending operation directly produce the effect (Johnson, P.S., 2001).

In the second mechanism, the elastomer is deformed beyond its breaking strain and fractures. In this fractured state, it mixes with the filler agglomerates and as it passes out of the high strain region, once again it seals the agglomerates inside (Johnson, P.S., 2001).

2.4.2 Dispersion

During dispersion, these agglomerates are broken down to a size of less than 1µm. The final degree of dispersion of filler depends not only on the specific characteristics of fillers (morphology, surface activity) but also on the mixing conditions (time, temperature, total shear strain) and on properties of the rubber (molecular weight, molecular weight distribution, chemical structure) (Johnson, P.S., 2001).

2.4.3 Distribution

In addition to breakdown of filler agglomerates, dispersive mixing, it is important that at the end of the mixing cycle the entire batch should be homogeneous. That is, a sample taken from any part of the batch should contain each of the ingredients in the proportion designated by the formula or compound recipe (Johnson, P.S., 2001).

2.4.4 Plasticization

Plasticizers or internal lubricants are added to rubber compounds to modify their bulk viscosities. As there become more closely incorporated into the mix, their effect is felt as a slight reduction in viscosity. This is not always seen as a change in Mooney Viscosity as this is measured at a very low shear rate compared to those experienced by the compound in the subsequent processing operations of milling, calendaring, extrusion, or molding (Johnson, P.S., 2001).

2.5 Flow Behaviour on Mills

Tokita, N. and White, J.L., 1966, classified the mill behaviour of raw elastomers as falling into four regions. A schematic illustration of this in Figure 2.7 shows the effect of temperature on milling behaviour as temperature is increased. In addition to temperature, they showed that the mill behaviour depends on the nip gap between the rolls, the ration of roll speeds, the actual roll speeds and the viscoelastic properties of the elastomer. They found that only at the region 2 did elastomers give good processing behaviour and accept fillers, oils, and chemicals readily.

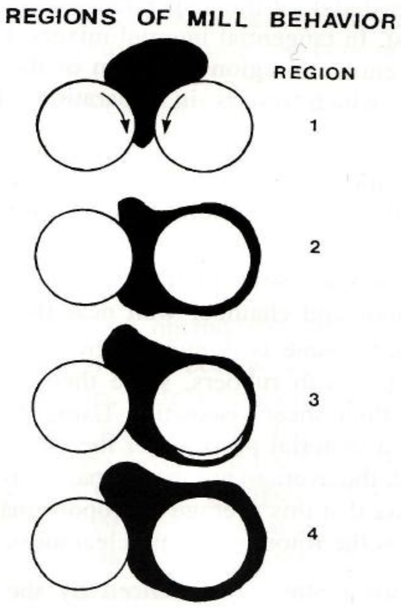


Figure 2.8: Effect of Temperature on Milling Behaviour of Rubber on a Mill

2.6 Effect of Compounding Ingredients on Processing Behaviour

The primary ingredient affecting the flow behaviour of a rubber compound is the base elastomer, or used elastomers. The choice is determined mainly by the end application, that is by the physical properties and environmental resistance required. However, rubber compounds contain in addition to elastomers, as many as ten or a dozen other ingredients in various proportion such that the elastomers comprises only 30 to 50% of the total weight. Other ingredients and their weight percentage in the formulation are deliberately chosen in order to affect the viscoelastic properties of the compound during subsequent processing steps. They are added to reduce frictional forces between rubber and metal. The remainder, added for some other purpose, may also inadvertently affect flow properties of the compound (Johnson, P.S., 2001).

2.7 Vulcanization process

Vulcanization is a process where polymer chains are converted to stable solid form from viscous form by cross linking these polymer chains. This process has a major effect on the properties of the polymer where a three dimensional elastic network would be created to produce useful mechanical properties (Dick, J.S., 2001).

Sulphur is the main component in this process of forming the cross linking, which depends on the presence of unsaturated bonds in the polymer chains. Physical properties of the vulcanizate depend on the cross linking structure and the type of the main chain. Other vulcanizing agents are used along with sulphur, such as accelerators, activators and retarders to balance the cure characteristics of the product (Faisal, M. A., 2005).

Accelerators are used to increase the cure rate while retarders delay the cure rate or the initiation of the cure. Activators are used to form complexes to speed up the reaction. (Faisal, M. A., 2005)

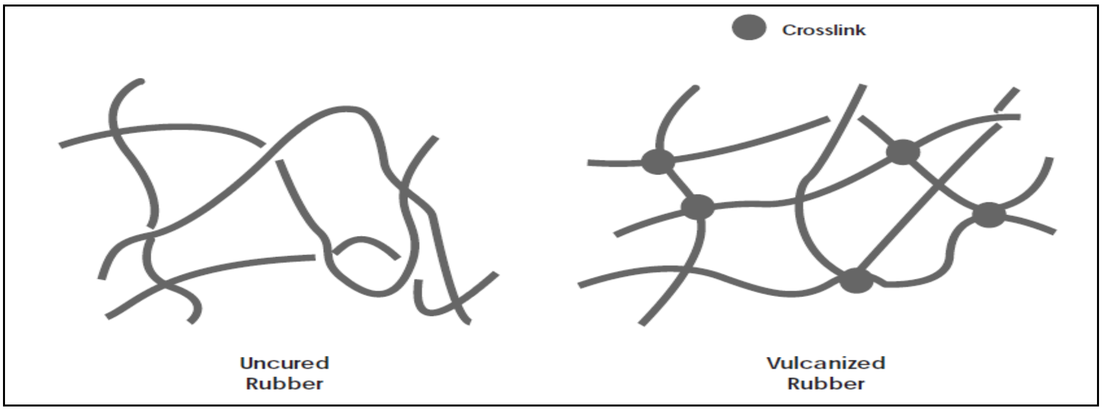


Figure 2.9: Uncured and Vulcanised Rubber

2.8 Molding

Many rubber articles are producing by molding, a process which uncured rubber is cured under pressure in a mold. A high viscosity compound may not fill the mold properly or may generate too much shear heat, leading to overheating or scorch. These considerations mean that there is a process window within which a specific material can be satisfactorily molded in given process.

Natural rubber and synthetic elastomers are commercially available in different viscosity grades. Lower viscosity elastomers are often chosen to ensure easy flowing compounds in the manufacture of molded goods. Higher viscosity grades are often used in extrusion grades because they provide high green-strength, even when highly extended with fillers and oils. However, as has been stressed earlier, the primary consideration in choosing a type and grade of elastomer is that it should provide the properties required in the end application (Johnson, P.S., 2001).

2.8.1 Compression Molding

In compression molding, Figure 2.10, the weight, dimensions, and positioning of the charge have to be closely controlled, or the dimensions of the product can vary widely. This is the often due to variation in the amount of material lost from the cavity as flash. This can be controlled, rather than be eliminated, by employing a shallow plunger, which, because of close clearances, means that excess rubber can only escape when high pressure is applied, after the mold is completely filled (Sommer, J., 1985).

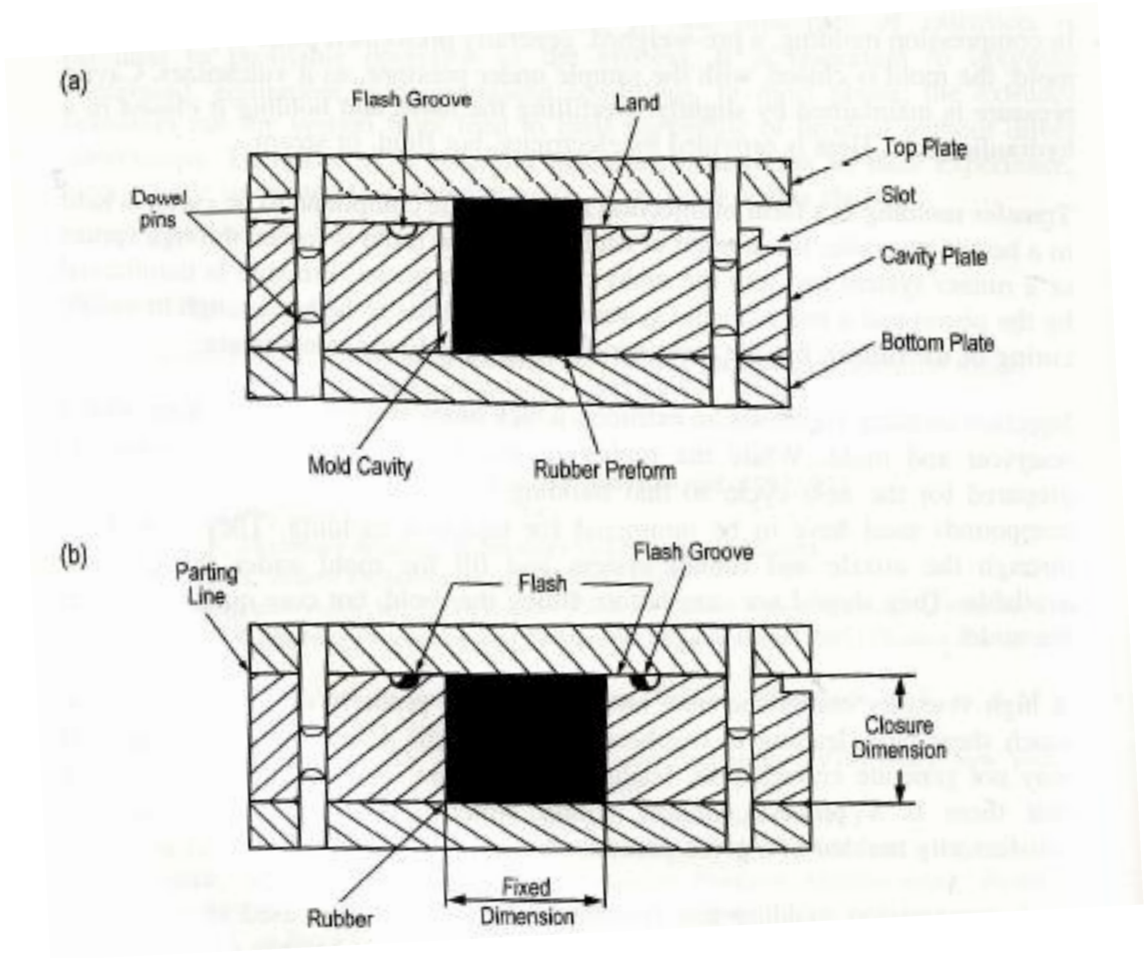


Figure 2.10: Compression Mold Containing Rubber Perform (a) Before Closing Molding And (b) After Mold Closing (Sommer, J., 1985).

2.9 Testings

2.9.1 Tensile test

Rubber is rarely used in simple tension, and never in practice is it stretched to be breaking point. However, one of the common standard tests involves stretching dumbbell shaped specimens and measuring the forces at 100 and 300% elongation at break. Tensile stress are, by convention, calculated based on the original cross section and are often referred to in the industry, incorrectly, as 100 and 300% modulus. The elongation at break is usually termed the ultimate elongation. Tensile properties are sensitive to the amount of diluents fillers, added to reduce volume costs, and more importantly, to poor filler dispersion or inadequate curing. (P.S. Johnson, 2001)

The effect of coconut filler content on maximum tensile stress, strain, and modulus of elasticity are shown in Figure 2.11. The increase of the filler content, results in the increase in tensile stress and tensile modulus of elasticity. This is due to the fact that coconut filler particles strengthen the interface of resin matrix and filler materials. The maximum tensile strength for 15% filler composite was higher (35.48 MPa) compared to other two combinations (Sapuan, S.M., et. al, 2003).

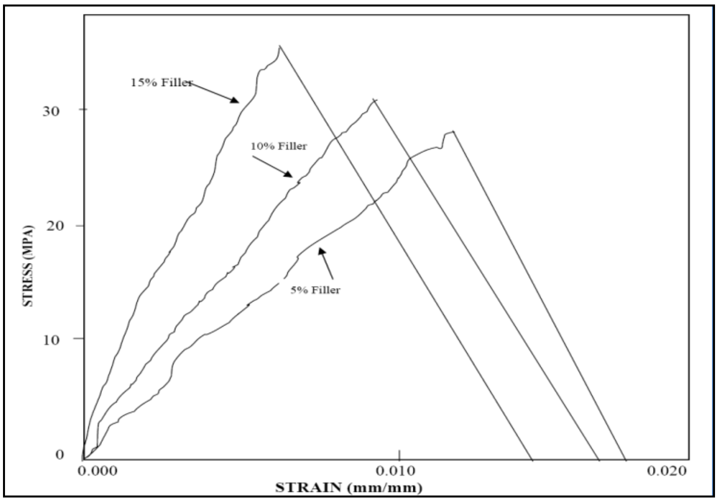


Figure 2.11**:** Stress versus Strain

Other methods to test Tensile properties of the vulcanizates were measured with a Monsanto tensile Tester (model VJ) at a cross-head speed of 500 mm min^{-1} using dumb bell test specimens (Sapuan, S.M., et. al, 2003).

Besides that, Tensile strength (TS), tensile modulus (E) and elongation at break (e) were determined according to ASTM D3039, under ambient conditions, using an Instron 5500R Universal Testing Machine (Instron Corp., Canton, MA) (Rosa, M. F., et.al, 2009)

Other source stated that the text specimens were molded in an electrically heated press at conditions predetermined from the rheographs. Tensile properties of the vulcanizates were measured with a Monsanto Tensile Tester (Mode 1/m) at crosshead speed of 500 mm/min using dumb bell test specimens (type H) as contained in ASTMD-412-87. The tensile strength at break was calculated as the elongation at break (Egwaikhide, A.P., et.al, 2008).

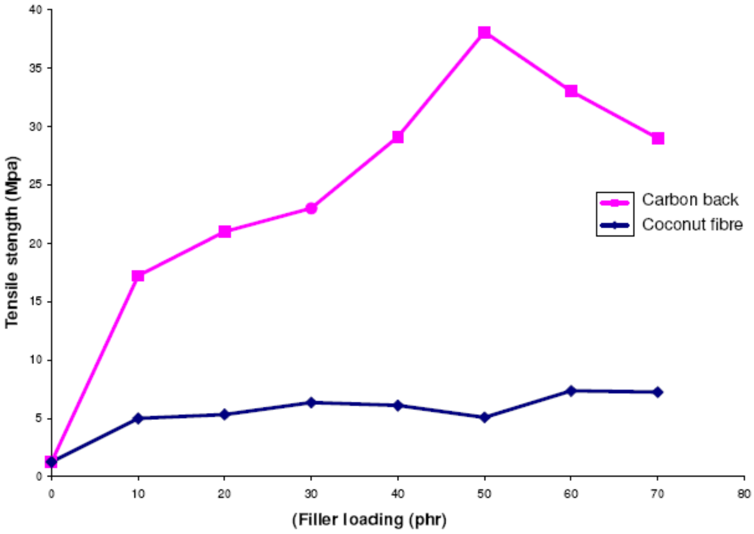


Figure 2.12**:** The effect of filler loading on tensile strengths of coconut fibre and carbon black N330.

The reinforcement of coir fibre in natural rubber has been extensively studied. Upon incorporation of coir fibre, it was seen that the tensile strength decreased sharply with increase in fibre loading up to 30 phr and then showed a slight increase for composites containing 40 and 60 phr fibre loading. This trend was observed in both longitudinal and transverse directions.

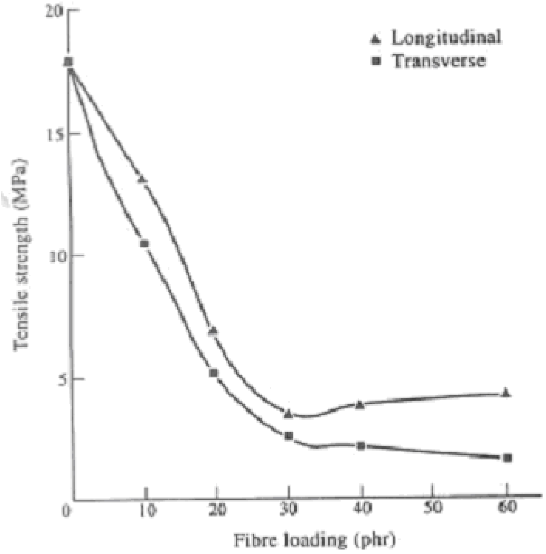


Figure 2.13: Variation in tensile strength with fiber loading.

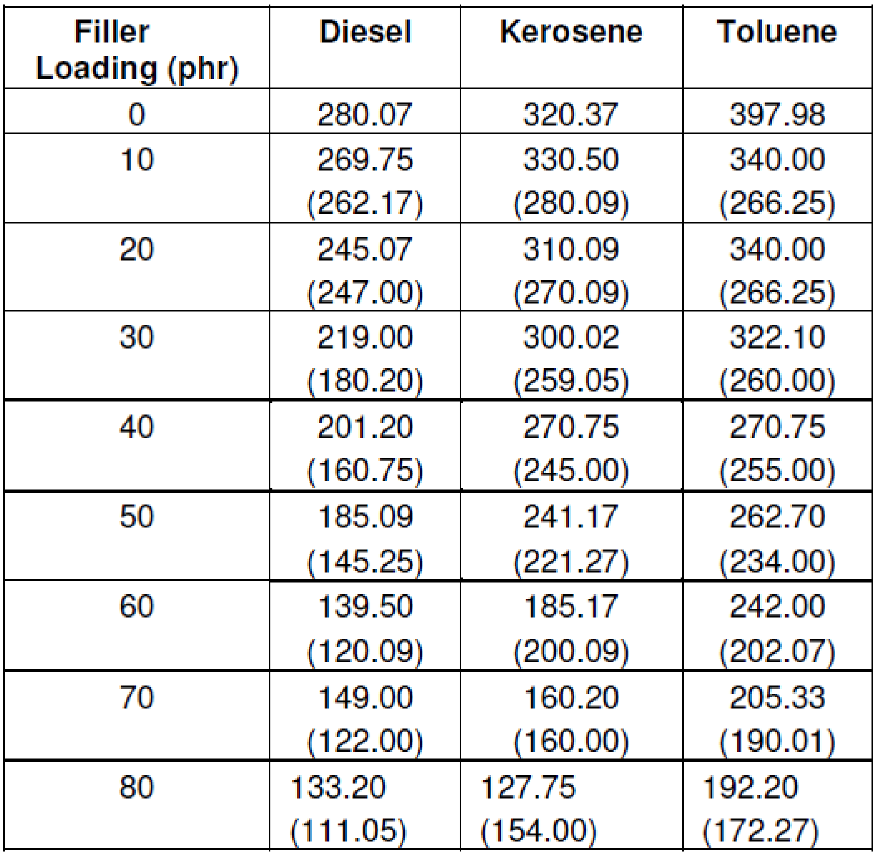
2.9.2 Swelling Test

In some application exposure to fluids, which could have adverse effects on rubber, occurs. Typical fluids such are gasoline, oil, air-conditioning solvents, antifreeze solutions, and aerosol propellants. The adverse effect that could occur are swelling of the rubber due to absorption of the fluid, extraction of some constituent such as plasticizer or antidegradants, or chemical reaction with the rubber or one of the compounding ingredients. The specific test applied depends on the nature of the solvent and expected effect on the rubber. The most generally applied test is to measure swelling after immersion in the liquid, for a specific time at a specific temperature. Swelling is usually measured by determining the volume change. It is common to measure also the effect of the immersion on tensile properties and hardness (Johnson, P.S., 2001).

The resistance of the vulcanizates to diesel, kerosene and toluene was determined by using the method described in ASTM-D3010. Three different shapes of the cured samples were cut from the 1mm thickness mould and weighed and immersed in air tight containers of diesel, kerosene and toluene solvents at ambient temperature for 24 hours. The samples were then removed from the bottles wiped dry with filter paper and weighed immediately. The change in weight of the sample was expressed as percentage swelling (Egwaikhide, P.A, et.al, 2008)

$$\%Swelling = \frac{W_2 - W_1}{W_1} \times 100$$

Table 2.4: Equilibrium Sorption in Diesel, Kerosene and Toluene of Natural Rubber Vulcanizate Filled with Coconut Fibre and Carbon Black (N330) (Egwaikhide, P.A, et.al, 2007).



Filler Loading (phr)	Diesel	Kerosene	Toluene
0	280.07	320.37	397.98
10	269.75 (262.17)	330.50 (280.09)	340.00 (266.25)
20	245.07 (247.00)	310.09 (270.09)	340.00 (266.25)
30	219.00 (180.20)	300.02 (259.05)	322.10 (260.00)
40	201.20 (160.75)	270.75 (245.00)	270.75 (255.00)
50	185.09 (145.25)	241.17 (221.27)	262.70 (234.00)
60	139.50 (120.09)	185.17 (200.09)	242.00 (202.07)
70	149.00 (122.00)	160.20 (160.00)	205.33 (190.01)
80	133.20 (111.05)	127.75 (154.00)	192.20 (172.27)

CHAPTER 3

METHODOLOGY

3.1 Introduction

For this study the type of method that is used is experimental method. This chapter is describes the details of processing of the composite. Besides that, the experimental procedure, apparatus and equipments that been applied for the purpose of this study is also been discussed.

3.2 Raw Materials

3.2.1 Natural Rubber (NR)

The natural rubber used in the experiment is Standard Malaysian Rubber-10 (SMR10). Natural or crude rubber is basically tough and an elastic solid. It becomes soft and sticky only as the temperature rises. Raw natural rubber has low tensile strength and is abrasion resistant. It absorbs large quantities of water. Its specific gravity is 0.915. The most important property of natural rubber is its elasticity. When stretched it expands and when released it attains its original state. This is due to its coil-like structure. The molecules straighten out when stretched and when released they coil up again. Therefore applying stress can easily deform rubber. But when this stress is removed it retains back its original shape. It is insoluble in water, alcohol, acetone, dilute acids and alkalis. It is soluble in ether, carbon disulphide, carbon tetrachloride, petrol and turpentine and other hydrocarbons like benzene, toluene, gasoline and lubricating oils. Natural rubber is basically a thermoplastic but with vulcanization it turns to thermoset.

3.2.2 Natural Fiber

3.2.2.1 Coconut Fiber

Coconut fiber is the filler to reinforce the natural rubber. Fillers range from inert dilatants such as whiting, talc, clays, $CaCO_3$, etc. which "cheapen" the product and make it go further, to reinforcing fillers which increase tensile strength and abrasion resistance. Carbon black is a most important reinforcing agent in tyres and tubes and is usually produced by burning oil or natural gas in a limited supply of oxygen. It is imported and it makes up to 30% of the tyre composition. For white or coloured products, silica, china clays and light magnesium carbonate are useful reinforcing agents, but inferior to carbon black in abrasive resistance. Fibrous fillers such as cotton flock, asbestos and synthetic resins can be used for special purposes. Nowadays natural fillers such as empty fruit bunch, palm oil fiber, coconut fiber, rice husk is been using to replace the synthetic fibers.

3.2.3 Activators

3.2.3.1 Zinc Oxide

Zinc oxide is an inorganic compound with the formula ZnO. It usually appears as a white powder, nearly insoluble in water. The powder is widely used as an additive into numerous materials and products including plastics, ceramics, glass, cement, rubber (e.g., car tires), lubricants, paints, ointments, adhesives, sealants, pigments, foods (source of Zn nutrient), batteries, ferrites, fire retardants, first aid tapes, etc. ZnO is present in the Earth's crust as the mineral zincite; however, most ZnO used commercially is produced synthetically.

Figure 3.1: Zinc Oxide

3.2.3.2 Stearic Acid

Stearic acid (first syllable pronounced either steer or stair) is the saturated fatty acid with an 18 carbon chain and has the IUPAC name octadecanoic acid. It is a waxy solid, and its chemical formula is $C_{18}H_{36}O_2$, or $CH_3(CH_2)_{16}COOH$. The salts and esters of stearic acid are called stearates.

Stearic acid is useful as an ingredient in making candles, plastics, dietary supplements, oil pastels and cosmetics, and for softening rubber. It is used to harden soaps, particularly those made with vegetable oil. Stearic acid is used in aerosol shaving cream products.

Figure 3.2: Stearic Acid

3.2.4 Antioxidant

3.2.4.1 N-(1,3-Dimethylbutyl)-N'-Phenyl-P-Phenylenediamine (6-PPD)

6PPD is used as an antioxidant and an antiozonant for natural and synthetic elastomers, as a stabilizer of polymers and as a monomer polymerization inhibitor. 6PPD is not regulated for use in food contact applications. 6PPD imparts high temperature stability, high strength, and chemical and electrical resistance. Antioxidant is a substance added in small quantities to hydrocarbons which are susceptible to oxidation, such as rubbers, plastics, foods, and oils to inhibit or slow oxidative processes, while being itself oxidized.

Figure 3.3: N-(1,3-Dimethylbutyl)-N'-Phenyl-P-Phenylenediamine (6-PPD)

3.2.5 Accelerator

3.2.5.1 N-Cyclohexyl-2-Benzothiazole Sulfenamide (CBS)

CBS is an accelerator with high activity and greatly delayed action. CBS is mainly used in manufacture of tires, rubber shoes, rubber tube, cable and other technical rubber goods. The properties of CBS are has a slight odour and non poisonous. Besides that, it soluble in benzene, methylbenzene, chloroform, carbon disulfide, carbon tetrachloride, methylene chloride, acetone and ethyl acetate and but not easily soluble in ethyl alcohol. CBS is insoluble in water, dilute acid, dilute alkali and gasoline.

Figure 3.4: N-Cyclohexyl-2-Benzothiazole Sulfenamide (CBS)

3.2.6 Vulcanizing Agent

3.2.6.1 Sulphur

Sulphur is the chemical element that has the atomic number 16. It is denoted with the symbol **S**. It is an abundant, multivalent non-metal. Sulfur, in its native form, is a bright yellow crystalline solid. In nature, it can be found as the pure element and as sulfide and sulfate minerals. It is an essential element for life and is found in two amino acids: cystiene and methionine. Elemental sulfur crystals are commonly sought after by mineral collectors for their brightly colored polyhedron shapes. Sulfur reacts directly with methane to give carbon disulfide, which is used to manufacture cellophane and rayon. One of the direct uses of sulfur is polymers. Sulfites are heavily used to bleach paper. Sulfites are also used as preservatives in dried fruit. Many surfactants and detergents, e.g. sodium lauryl sulfate, are produced are sulfate derivatives.

Figure 3.5: Sulphur

3.3 Equipments

3.3.1 Two Roll Mill

Two roll mill consists of two horizontal, parallel, heavy metal rolls which can be jacketed with steam and water to control temperature. These rolls turn towards each other with a pre-set, adjustable gap or nip to allow the rubber to pass through to achieve high shear mixing. The back roll usually turns faster surface speed than the front roll; the difference increases shear forces. The difference in roll speeds is called the “friction ratio”. Mill mixing is the oldest method of rubber mixing, relatively slow method and its batch size is limited. The model of laboratory two roll mill used to do the experiment is Lotus LS 22005.

Figure 3.6: Two roll mill

3.3.2 Hot Molding Press

LS 22025 ton hot and cold molding press used to mold the sample. Compression Molding is the original production method for molded rubber. It is ideal for low to medium volumes and a useful molding process for forming bulky parts, gaskets, seals and O-rings. It is a widely used, efficient, economical production method for many products particularly low production volumes of medium to large parts, materials with a high cost and applications that demand extreme hardness. It takes place as two plates of mold are forced together. A slightly oversized rubber preform (raw compound) is shaped to approximately fit into the mold cavity. When the plates are squeezed together, the preform is compressed and forced to conform to the mold cavity. A molding press is used to provide the necessary force to close the mold. Excess material, or flash, that squeezes out is pushed into grooves cut into the top plate and later trimmed off. Once the curing or vulcanizing process is complete, the mold is opened and the molded product can be removed).

Figure 3.7: Hot and Cold Molding Press

3.3.3 Universal Tensile Tester

Instron 3369 model of universal tensile tester with 50 kN load has been used to test the strength of the samples. Tensile tests measure the properties of tensile strength. The standardized test piece is held in the testing machine and an increasing tensile force is applied until the material breaks down. The results of any testing are produced as a graph which shows the relationship between force and extension, and also the nominal stress against the strain. If a small force is applied the extension is proportional to the force. Further force causes the sample to stretch rapidly. "Necking" or "wasting" occurs shortly after the maximum force U has been reached and just before final fracture occurs

Figure 3.8: Universal Tensile Tester

3.4 Formulation of Composite

Table 3.1: Recipe for General Compounding the Natural Rubber

INGREDIENTS	PHR (PARTS PER HUNDRED)
SMR-10	100.0
Coconut fiber	0,15,30,40,50
Stearic Acid	2.0
Zinc Oxide	5.0
6-PPD	1.0
CBS	0.5
Sulphur	2.5

3.5 Method Of Research

3.5.1 Sample Preparation

3.5.1.1 Fiber Preparation

The coconut husk is bought from supplier. Then the husk is grinded using hammer mill to make it in a powder form. Then the powder is sieved manually using 630µ sieve plate to obtain the desired coconut fiber. During sieving process, safety precautions need to be considered. Wearing mask, gloves and goggle is essential to avoid any health effect.

Figure 3.9: Coconut Husk before Grind

Figure 3.10: Sieving Process

Figure 3.11: Coconut Fiber

3.5.1.2 Rubber Preparation

Natural rubber is cut using saw from the bulk SMR10 block. Then the rubber is weight according to the formulation given either for 0, 15, 30, 40, and 50 parts of fillers. The detailed formulation is attached in appendix.

Figure 3.12: Raw Natural Rubber (SMR10)

Figure 3.13: SMR10

3.5.2 Procedures

3.5.2.1 Part 1

The rubber and coconut fiber is weighed according to the recipe calculated. The main switch of two roll mill is turned on and the switch on the panel also be turned on. Then the heater and motor is turned on. The temperature is set to be 80^0C. The speed of motor can be varies depends on the sample. Then the rubber is rolled in two roll mill to get a thin layer. Then the fiber is added in a small amount first as it covers the rubber and make a coating of coconut fiber on top of it. Then the rubber is fold in two and blend it again until the composite becomes thin again. Then the filler is added again. The same steps repeated until all the fiber is used and the composite becomes homogeneous. The time to mix the composite is depends on the filler loading. For the mixing process, it is advised not to use gloves as it can be stick to the sample and bring injury if there is any lack of safety precaution.

Figure 3.14: Rubber with Coconut Fiber

3.5.2.2 Part 2

Stearic acid, zinc oxide and 6-PPD are weighed according to the recipe. The set point of temperature for two roll mil is set about 50^0C. Then the composite of natural rubber and filler is blended with the chemicals using the two roll mill. The composite need to be homogenous as it will be rolled for few times. The time taken for mixing is about 10 minutes.

3.5.2.3 Part 3

The blended composite is needed to be cured before molded. Sulphur and CBS are weighed followed the recipe. Using the same temperature from part two, the chemicals are blended together with the composite again using two roll mill until the reinforced rubber reaches homogenous. The temperature needed to blend sulphur and CBS is about 50^0C and the time required to mix them is around 10 minutes.

Figure 3.15: Reinforced Rubber

3.5.2.4 Molding

The reinforced rubber is molded using hot press machine with temperature 160^0C and left it in the press for 30 minutes. The reinforced rubber is cut to small pieces and placed it between two metal molding plates. The main switch of hot press is on and the heater is turned on and the temperature is set to 160^0C. The machine is left until the temperature of hot plates rise to 160^0C. The sample which is between the metal molding plates is placed between on the lower part of hot plate in the hot press machine. Later, the pump and load buttons are turned on and let the shaft to attach with the upper hot plate and the pump is stopped. The timer is set to 30 minutes. After 30 minutes, the pump is turned on back and the unload button is press until the shaft lowers. Stop button is pressed and the hot mold plates are taken out. The mold is left in the room temperature to cool down before the molded rubber is separated from the mold. Sample for testing is cut manually from the molded rubber. For tensile test, the samples are in dumbbell shaped. The same procedure is used for different filler loadings.

Figure 3.16: Molded Reinforced Rubber

3.6 Testing

3.6.1 Tensile Test

Dumbbell-shaped samples were cut manually from the mold sheets. Dimension such as width, length, and thickness of the specimens are jotted down as need to key in the data in the software. Tensile properties were determined using Instron 3369 Universal Testing Machine with a crosshead speed of 20 mm/min while the load used is 50 kN. The software used to obtain the data is Bluehill. Five samples were tested for each filler loading.

First, grip the sample tightly at the clamp in tensile tester. Then the required testing that wanted to be tested is key in the software. Other essential information also has been key in the software. Next, the load needs to be balanced before the test start. Later, press the start button to begin the testing. After the break of specimen, the machine will stop automatically and specimen needs to be removed. Lastly, the results that have been calculated need to be saved for the further analysis.

Figure 3.17: Dumbbell Shaped Sample

Figure 3.18: Initial Specimen

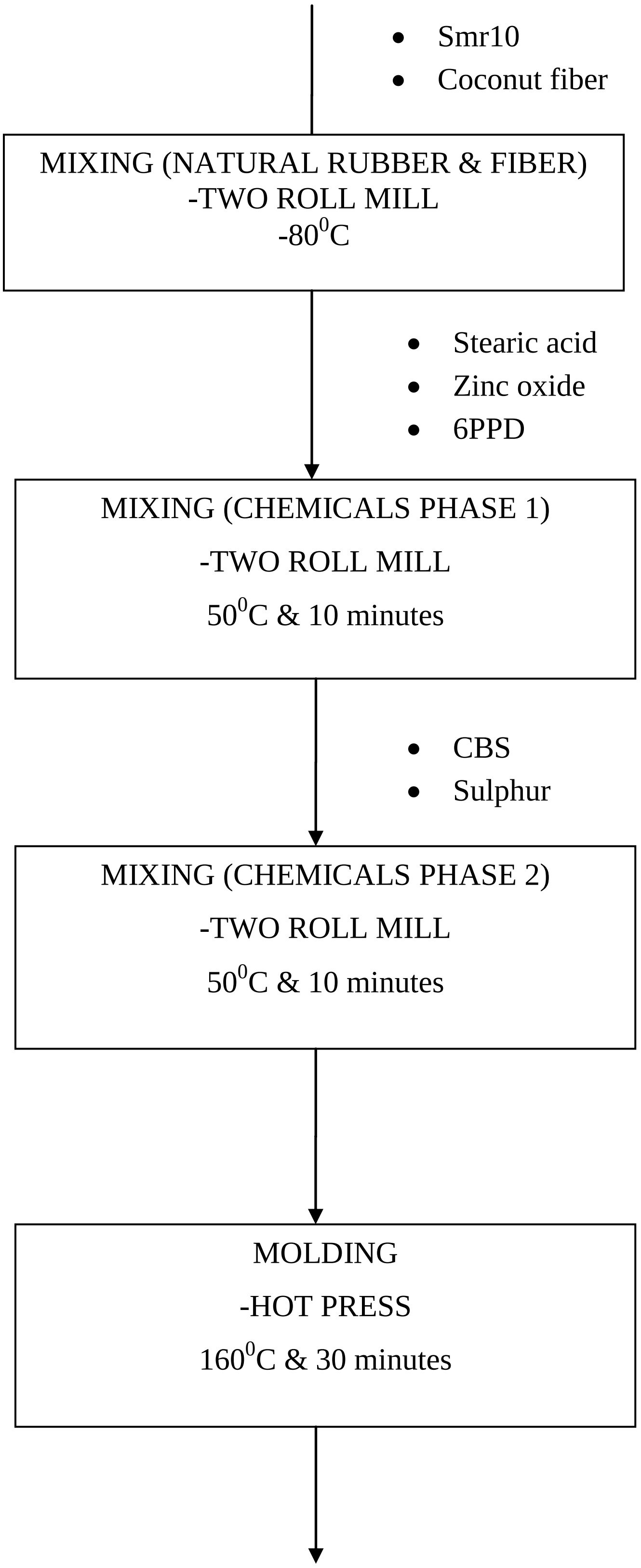
Figure 3.19: During Elongation

Figure 3.20: Final Specimen after Break

3.6.2 Swelling Test

Two specimens for each filler loading are cut from the molded rubber for swelling test. Then the weight of each specimen is weighed and recorded. The samples are immersed in the different solvents such as kerosene and diesel in a 250ml schott bottle. All the bottles need to be labeled to avoid any confusion. The time recorded after the all the specimens are been tighten up in the bottle. Later, after 24 hours the sample is taken out from the solvents and wiped with filter paper to remove excess solvent. Then the weight of each sample is recorded again. Then using the swelling formula, the percentage of swelling is determined. Gloves, apron, mask and goggle need to be wear during handling with the solvents.

Figure 3.21: Swelling Test Samples


Smr10
Coconut fiber
MIXING (NATURAL RUBBER & FIBER)
-TWO ROLL MILL
-80^{0}C
Stearic acid
Zinc oxide
6PPD
MIXING (CHEMICALS PHASE 1)
-TWO ROLL MILL
50^{0}C & 10 minutes
CBS
Sulphur
MIXING (CHEMICALS PHASE 2)
-TWO ROLL MILL
50^{0}C & 10 minutes
MOLDING
-HOT PRESS
160^{0}C & 30 minutes

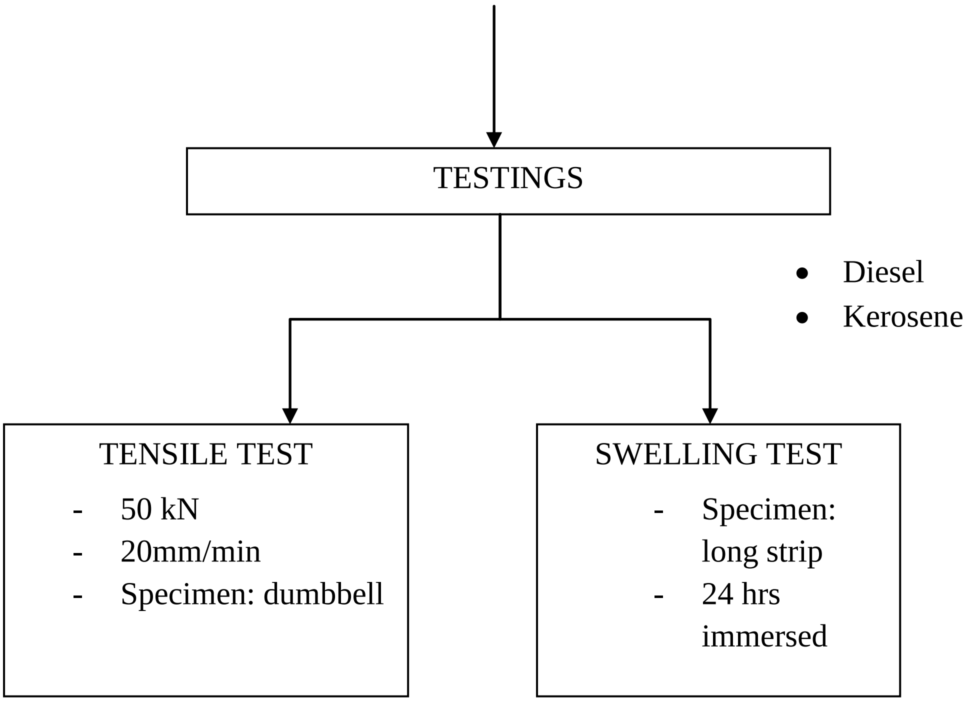


Figure 3.22: Flow Chart of Research Methodology

CHAPTER 4

RESULTS AND DISCUSSIONS

4.1 Introduction

The result of this study is discussed in this chapter. The tests such as tensile test and swelling test has been done to analyze the properties of natural rubber which has been added with fillers and other additives. Besides that, the virgin natural rubber will be used to compare the data. Moreover, the analysis on mixing also discussed in this chapter. There are graphs that assist the analysis on the properties of rubber.

4.2 Stress versus strain

Figure 4.1 shows graph of stress versus strain for reinforced rubber. It can be concluded that different filler loading to the natural rubber, it gives the different value of stress and strain. Based on theory, unlike metals, the stress-strain curve for typical rubber compound has no linear portion. Therefore it is usually not practical to calculate Young's modulus, which would be thee slope of a straight line drawn tangent to the curve and passing through the origin. Instead, stress at selected elongation is usually reported (Dick, J.S., 2001).

Stress-strain properties, such as ultimate tensile strength can be easily affected by poor mixing and dispersion, the presence of contamination, under-curing, over-curing and porosity, among other factor. Undispersed particles of different compounding ingredients, such as fillers, cause stress concentrations during the stretching of a rubber dumbbell, causing premature breaks at lower stresses. Impurities such as dirt also can cause the dumbbell to break in a lower stress. Likewise, volatile compounding ingredients can cause porosity to form during cure. These voids can cause lower tensile strength (Dick, J.S., 2001).

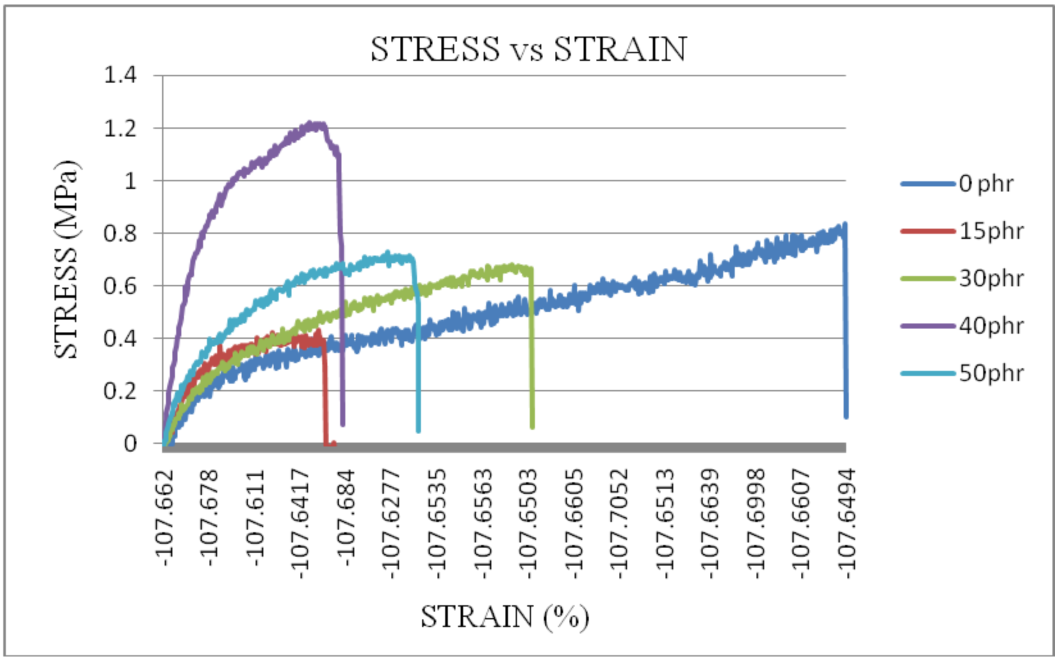


Figure 4.1: Stress versus Strain

4.3 Effect of filler loading towards tensile strength

From the results obtained, Figure 4.2 shows the graph of tensile strength versus filler loading. The tensile strength for the 15 phr loading is decreases to the maximum and increases gradually for 30 phr and 40 phr. Later, the tensile strength decreases for the 50 phr loading. Based on the theory, when the natural filler loading is increasing, the tensile strength will be decreasing.

According to Ismail, H., et.al, 2000, the tensile strength may increase or decrease when there is incorporation of fillers into polymer. The factor of evenness of filler will affect the tensile strength. The strength will normally decrease when the filler is unable to support stresses transferred from the polymer matrix.

Green strength is the tensile strength and/or tensile modulus of an uncured rubber compound (Dick, J.S., 2001). Besides that, it stated that the aspect ratio of lignocellulosic fibers did not necessarily result in the improvement of the strength of a composite, especially if there was widespread fiber agglomeration in the rubber matrix. As the filler loading is increased, eventually a level is reached whereby the filler particles or aggregates no longer adequately separated or wetted by rubber phase (Egwaikhide, P.A., et.al, 2007).

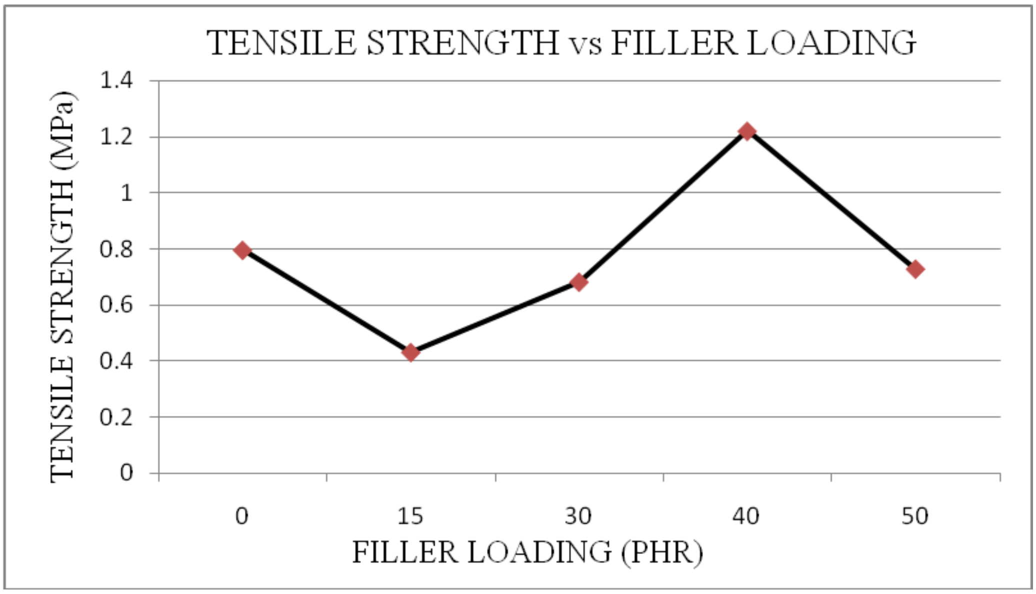


Figure 4.2: Tensile Strength versus Filler Loading

4.4 Effect of filler loading towards Extension at break

Figure 4.3 shows that the graph of extension at break versus filler loading. From the graph, the extension continues decreases from 0 phr, 15 phr and 30 phr. Later, for 40 phr and 50 phr, the extension increases. This may be due to poor dispersion of coconut fiber at higher level. The elongation affects by the stiffness achieved from strong adhesion between fiber and rubber matrix.

Elongation at break means the measure of ductility of the material determined from tensile test. The percent elongation is the percentage of strain at fracture or:

$$\%EL = \frac{L_f - L_0}{L_0} \times 100$$

Consequently, the toughness of the composite is reduced; resulting in lower elongation at break. Higher toughness is obtained from weak interfacial adhesion as shown by longer elongation at break for compound filled with fibers. Weak interfacial bond resulted in energy adsorption mechanism, i.e. bond breakage at the fiber/matrix interface (Ismail, H., et.al, 1997).

Ismail, H., et.al, 2000, also reported that the incorporation of natural fillers into natural rubber composites reduces the elongation at break. It is also reported that decreasing of elongation at break with addition of more filler to the polymer matrix. With increase in filler loading, the stiffness and brittleness of the composite increase gradually with an associated decrease in the elongation at break.

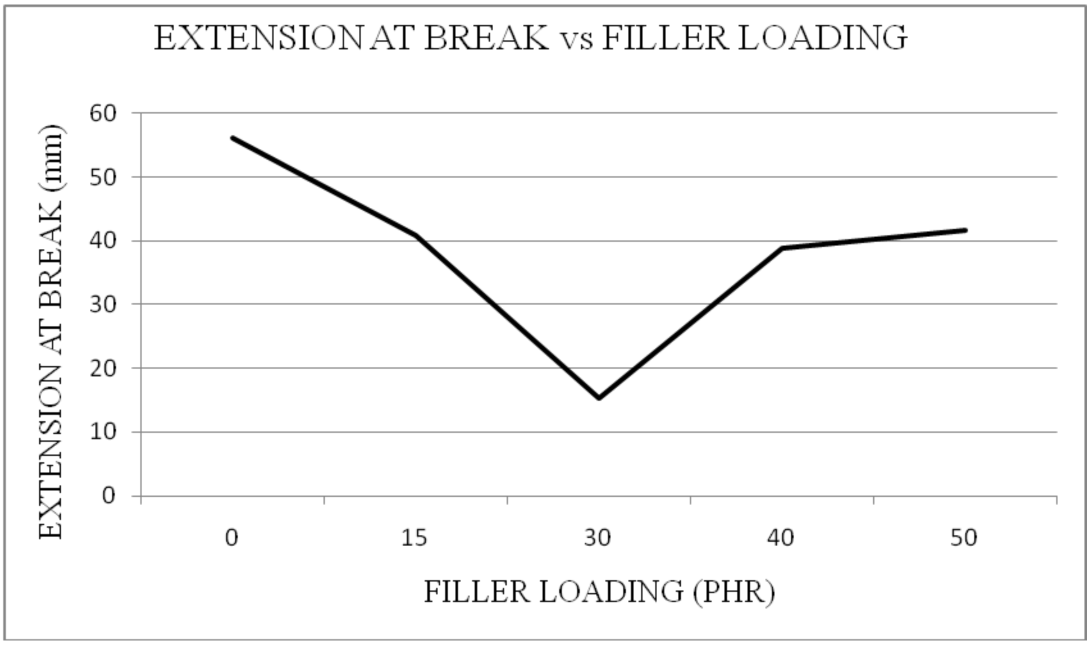


Figure 4.3: Extension at Break versus Filler Loading

4.5 Effect of Time at Break Due To Filler Loading

Figure 4.4 shows that graph of time at break versus filler loading. The time need to break the 0 phr filler to the rubber is longest due to the elasticity. The time at break decreases for 15 phr and increases for 30 phr. The increment of time for 30 phr may be due to the poor dispersion of coconut fiber to the rubber. Subsequently, for 40 phr and 50 phr, the time at break getting shorter.

The time at break depends on the stiffness of the composite. The more elastic the sample, the more time required to break the bond. One of the factors that can influence the result is curing process. It is because the sample may be over curing or under curing that will affect the elongation and indirectly effecting the time at break.

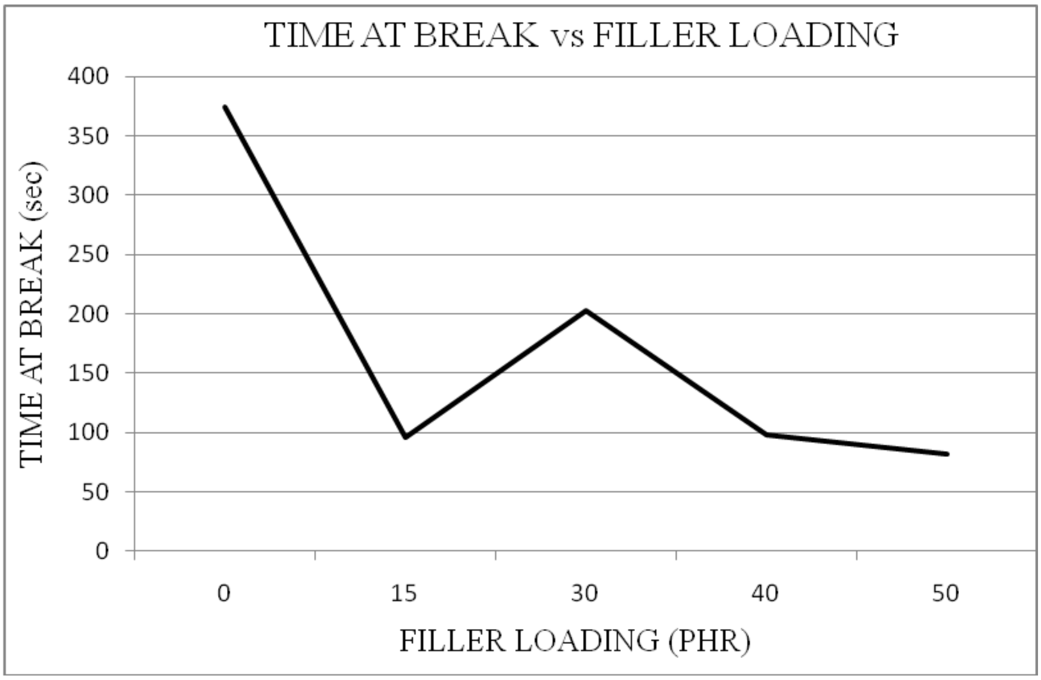


Figure 4.4: Time at Break versus Filler Loading

4.6 Effect of Filler Loading Towards Mixing Time

Figure 4.5 shows the graph of mixing time versus filler loading. From the graph, it can be concluded that the time required for mixing of fillers and chemical to the natural rubber is increasing as the filler loading increases. The primary factor that determines the mixing time is filler incorporation.

Johnson, P.S., 2001**,** has reported that the filler is compressed by the rubber and the agglomerates are crushed by shearing and compressive forces. As the filler loading increasing and the mixing process continue, the rubber is forced into the void in the filler and the air is expelled. When all the air is expelled and replaced by polymer the mix reaches its final density. Further distributive and dispersive mixing does not affect this, and the density does not change further once incorporation is complete.

The weight and volume of coconut fiber increases, while the weight of rubber and other additives decreases. As the volume of coconut fiber increases, the mixing process becomes tough.

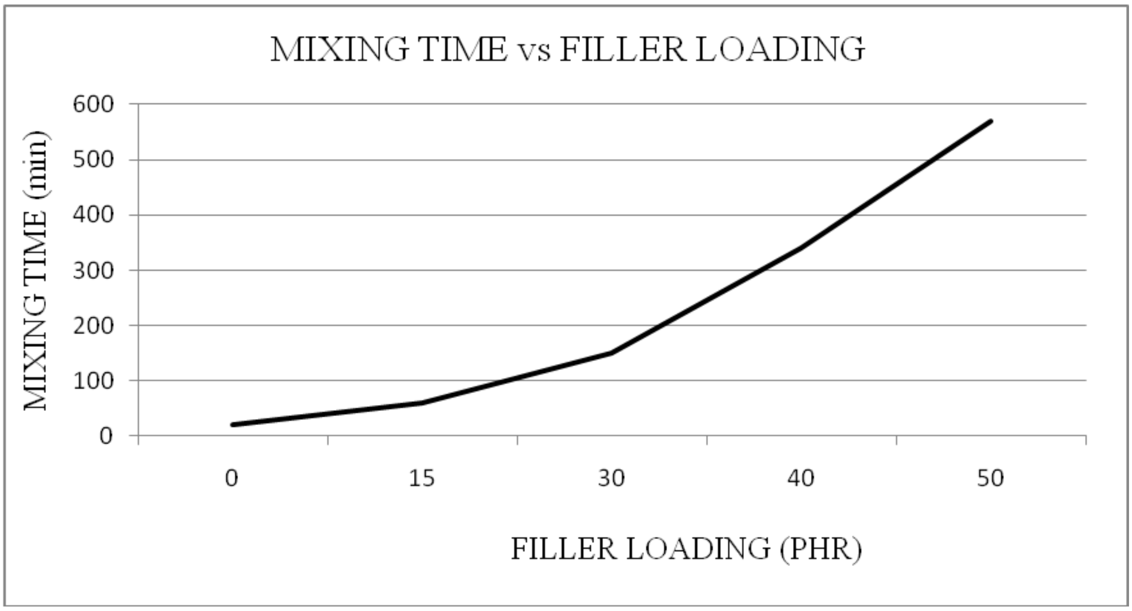


Figure 4.5: Mixing Time versus Filler Loading

4.7 Swelling Test

Figure 4.6 shows the graph of percentage swelling versus Filler loading. From the graph, it can be conclude that the percentage of swelling using solvents such as kerosene and diesel is decreasing as the filler loading is increasing. Within two solvents that have been used for this test, the cure rubber has the most swelling percentage when immersed in kerosene. It is because kerosene being a mixture of hydrocarbon (C10 – C14) with a lower molecular weight than diesel (C14 – C19) may be expected to diffuse faster and be accommodated in the rubber matrix.

Based Dick, J.S., 2001, liquid immersion properties are very important when developing a new rubber compound that may have contact with various liquids while in service. For example, rubber products, such as seal, may be exposed to an oil environment if used in an automatic engine. The ability of a cured rubber specimen to withstand certain liquids without swelling is highly relevant to the rubber product's service life.

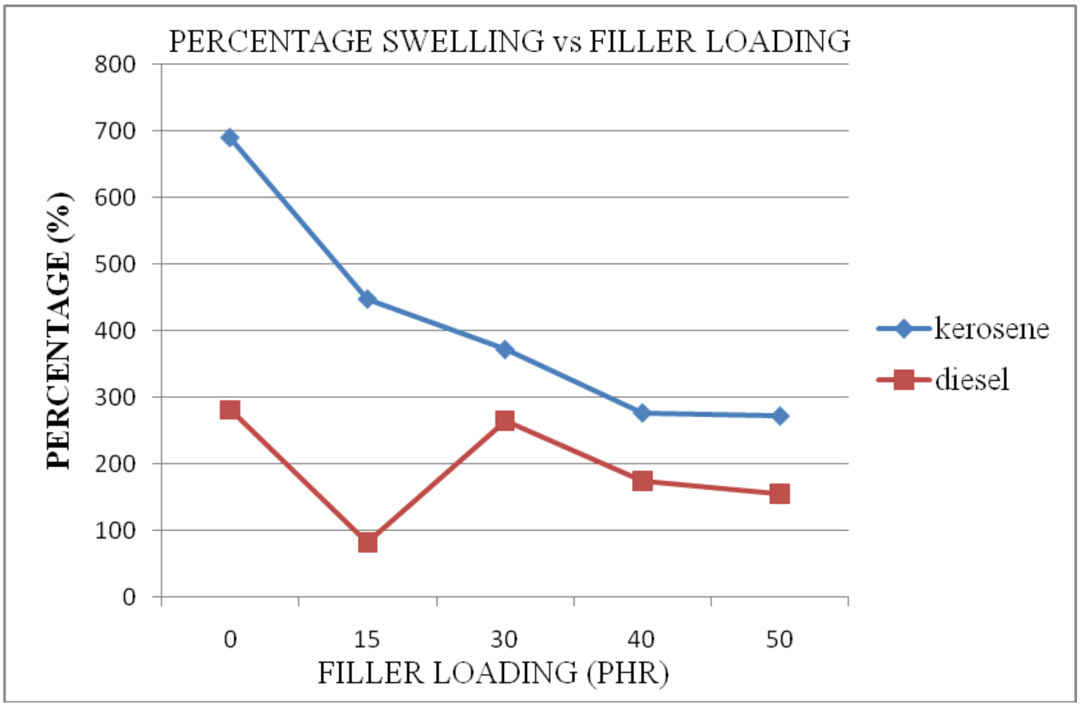


Figure 4.6: Percentage Swelling versus Filler Loading

CHAPTER 5

CONCLUSIONS AND RECOMMENDTIONS

5.1 Conclusions

The objectives have been achieved and there are few conclusions that can be made from this research. First of all, the stress-strain graph for the virgin and reinforced rubber has different value of stress and strain as the properties, such as ultimate tensile strength can be easily affected by poor mixing and dispersion, the presence of contamination, under-curing, over-curing and porosity.

Next, the tensile strength is decreases when the filler loading is increases. It is because evenness of filler will affect the tensile strength. The strength will normally decrease when the filler is unable to support stresses transferred from the polymer matrix. As the filler loading is increased, eventually a level is reached whereby the filler particles or aggregates no longer adequately separated or wetted by rubber phase.

Besides that, natural fiber has the most elongation and when the filler loading increases the elongation decreases along with the reinforced rubber becomes more stiff and hard. The elongation affects by the stiffness achieved from strong adhesion between fiber and rubber matrix. The incorporation of natural fillers into natural rubber composites reduces the elongation at break.

In addition, mixing time increases as the filler loading increase. It happens because the fillers getting harder to be distributed in the rubber. As the filler loading increasing and the mixing process continue, the rubber is forced into the void in the filler and the air is expelled.

For swelling test, it can be conclude that the more the filler loading increases in natural rubber, the swelling percentage is decreasing. Besides that, when the solvent has lower molecular weight with less carbon, it will diffuse more into the rubber and the rubber getting swelled.

5.2 Recommendations

There are few recommendations can be suggested to improve the research. First of all, the fibers that been sieved for the mixing process need to be make sure that the fibers are even in size. The evenness of fiber can affect the strength of the rubber.

Secondly, mixing of the natural rubber with other additives need to be more homogeneously distributed when using two roll mill. Better dispersion can increase the strength of rubber. Moreover, the speed and cleanliness need to be controlled well so that there won’t be any disturbances in the process. For that, it is recommended to use the internal mixer with high shear stress. It can save time and energy consuming as I can be more productive.

Next, the curing process needs to be assured whether it is correctly done. It is because if there are under-curing or over-curing, the mechanical properties of reinforced rubber will decrease and the result will not be as expected. In addition, the temperature is needed to be kept in minimum during curing process.

The samples for testing should follow the standard such as ASTM standard. For prepare the sample, it is suggested to use injection molding machine to mold the samples. By using the standards that has been fixed for rubber testing, the results that will be obtained are more accurate and reliable.

Lastly, the rubber that been reinforced with natural fillers is more suitable for the rubber products with less strength needed. This product is not suitable for production of tires because they need high strength and less swelling percentage.

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APPENDIX A

TABLES OF RUBBER FORMULATION

Ingredient	PHR	SG	Volume	Weight	Weight/250g
SMR-10	100	1.03	97.0874	100	225.23
Stearic Acid	2	0.85	2.3529	2	4.50
Zinc Oxide	5	5.57	0.8977	5	11.26
6-PPD	1	1	1.0000	1	2.25
CBS	0.5	1.3	0.3846	0.5	1.13
Sulphur	2.5	2.07	1.2077	2.5	5.63
Fillers	0				0.00
Total	111		102.9303	111	250.00

APPENDIX A1: Formulation for 0% Filler Loading

Ingredient	PHR	SG	Volume	Weight	Weight/250g
SMR-10	100	1.03	97.0874	100	198.41
Stearic Acid	2	0.85	2.3529	2	3.97
Zinc Oxide	5	5.57	0.8977	5	9.92
6-PPD	1	1	1.0000	1	1.98
CBS	0.5	1.3	0.3846	0.5	0.99
Sulphur	2.5	2.07	1.2077	2.5	4.96
Fillers	15				29.76
Total	126		102.9303	111	250.00

APPENDIX A2: Formulation for 15% Filler Loading

Ingredient	PHR	SG	Volume	Weight	Weight/250g
SMR-10	100	1.03	97.0874	100	177.30
Stearic Acid	2	0.85	2.3529	2	3.55
Zinc Oxide	5	5.57	0.8977	5	8.87
6-PPD	1	1	1.0000	1	1.77
CBS	0.5	1.3	0.3846	0.5	0.89
Sulphur	2.5	2.07	1.2077	2.5	4.43
Fillers	30				53.19
Total	141		102.9303	111	250.00

APPENDIX A3: Formulation for 30% Filler Loading

Ingredient	PHR	SG	Volume	Weight	Weight/250g
SMR-10	100	1.03	97.0874	100	165.56
Stearic Acid	2	0.85	2.3529	2	3.31
Zinc Oxide	5	5.57	0.8977	5	8.28
6-PPD	1	1	1.0000	1	1.66
CBS	0.5	1.3	0.3846	0.5	0.83
Sulphur	2.5	2.07	1.2077	2.5	4.14
Fillers	40				66.23
Total	151		102.9303	111	250.00

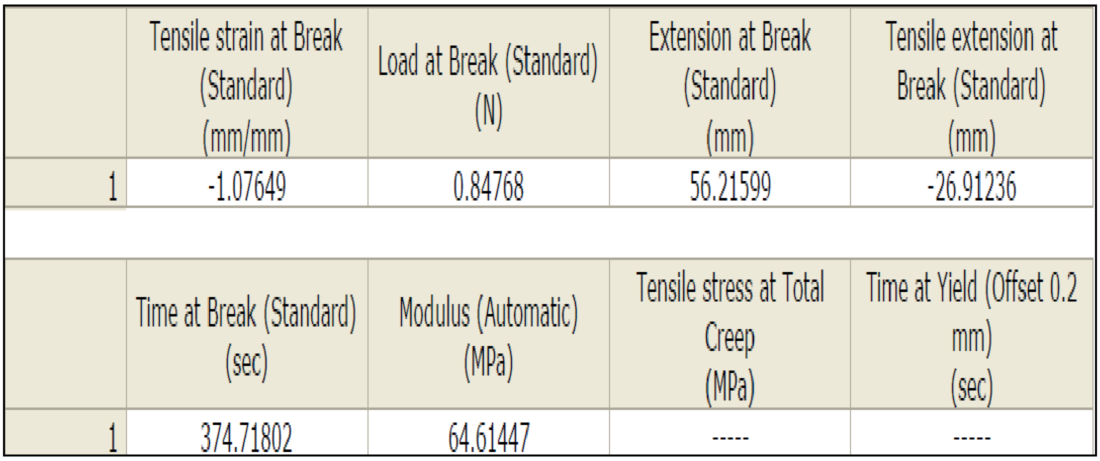
APPENDIX A4: Formulation for 40% Filler Loading

Ingredient	PHR	SG	Volume	Weight	Weight/250g
SMR-10	100	1.03	97.0874	100	155.28
Stearic Acid	2	0.85	2.3529	2	3.11
Zinc Oxide	5	5.57	0.8977	5	7.76
6-PPD	1	1	1.0000	1	1.55
CBS	0.5	1.3	0.3846	0.5	0.78
Sulphur	2.5	2.07	1.2077	2.5	3.88
Fillers	50				77.64
Total	161		102.9303	111	250.00

APPENDIX A5: Formulation for50% Filler Loading

APPENDIX B

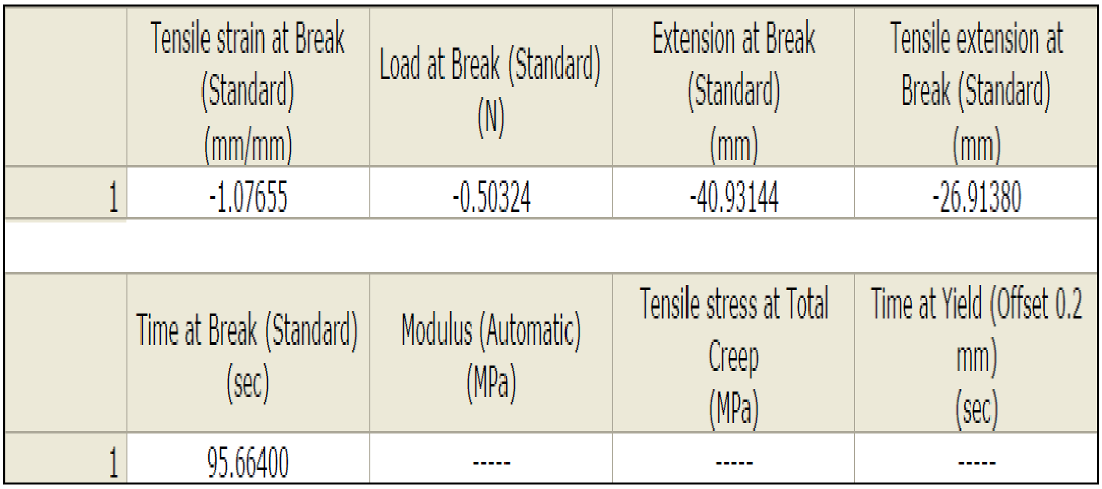
DATA FROM TENSILE TEST



	Tensile strain at Break (Standard) (mm/mm)	Load at Break (Standard) (N)	Extension at Break (Standard) (mm)	Tensile extension at Break (Standard) (mm)
1	-1.07649	0.84768	56.21599	-26.91236

	Time at Break (Standard) (sec)	Modulus (Automatic) (MPa)	Tensile stress at Total Creep (MPa)	Time at Yield (Offset 0.2 mm) (sec)
1	374.71802	64.61447	-----	-----

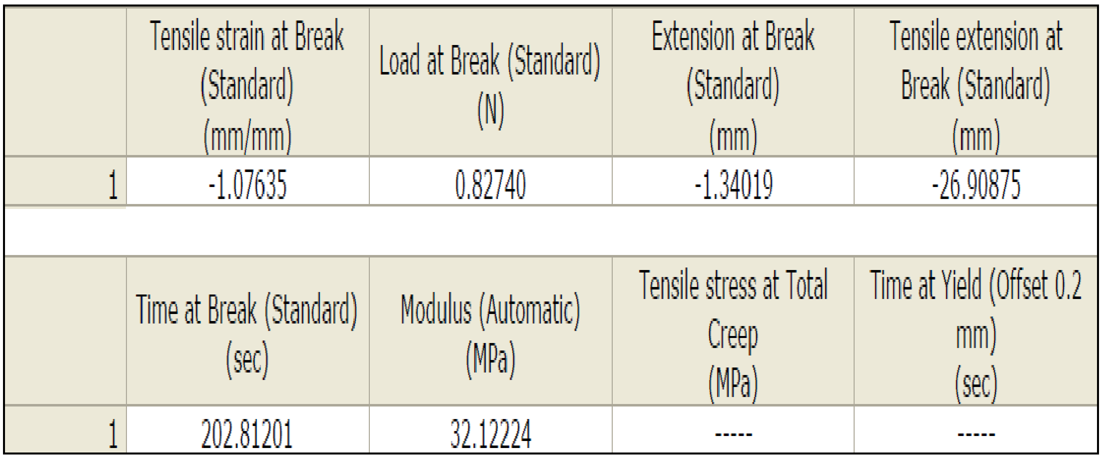
APPENDIX B1: Data of 0% Filler Loading



	Tensile strain at Break (Standard) (mm/mm)	Load at Break (Standard) (N)	Extension at Break (Standard) (mm)	Tensile extension at Break (Standard) (mm)
1	-1.07655	-0.50324	-40.93144	-26.91380

	Time at Break (Standard) (sec)	Modulus (Automatic) (MPa)	Tensile stress at Total Creep (MPa)	Time at Yield (Offset 0.2 mm) (sec)
1	95.66400	-----	-----	-----

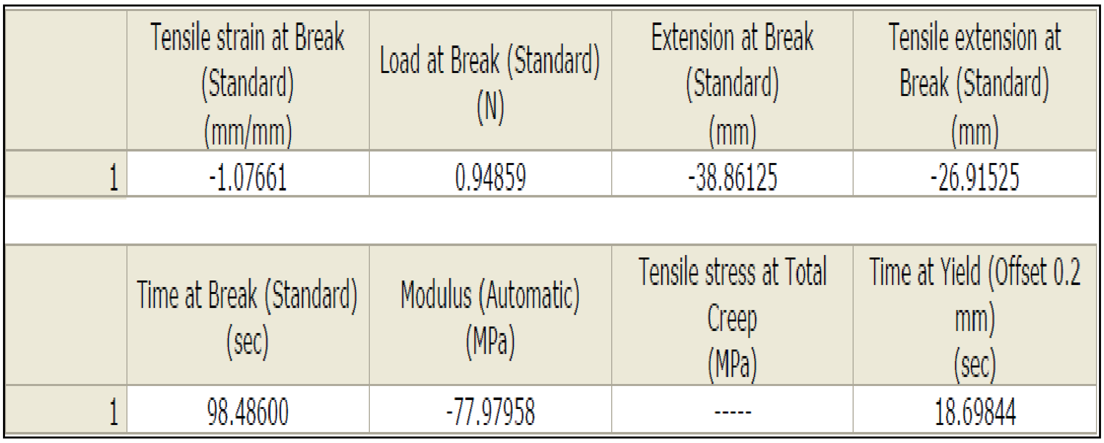
APPENDIX B2: Data of 15% Filler Loading



	Tensile strain at Break (Standard) (mm/mm)	Load at Break (Standard) (N)	Extension at Break (Standard) (mm)	Tensile extension at Break (Standard) (mm)
1	-1.07635	0.82740	-1.34019	-26.90875

	Time at Break (Standard) (sec)	Modulus (Automatic) (MPa)	Tensile stress at Total Creep (MPa)	Time at Yield (Offset 0.2 mm) (sec)
1	202.81201	32.12224	-----	-----

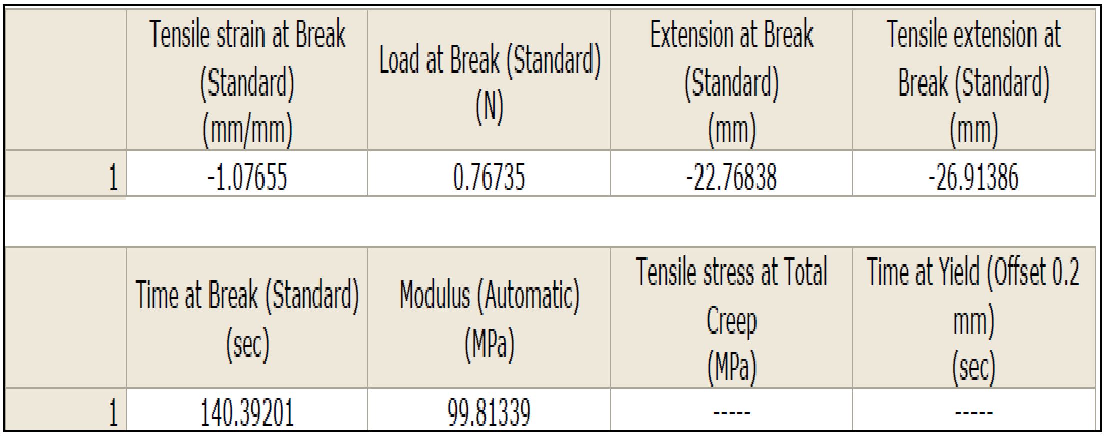
APPENDIX B3: Data of 30% Filler Loading



	Tensile strain at Break (Standard) (mm/mm)	Load at Break (Standard) (N)	Extension at Break (Standard) (mm)	Tensile extension at Break (Standard) (mm)
1	-1.07661	0.94859	-38.86125	-26.91525

	Time at Break (Standard) (sec)	Modulus (Automatic) (MPa)	Tensile stress at Total Creep (MPa)	Time at Yield (Offset 0.2 mm) (sec)
1	98.48600	-77.97958	-----	18.69844

APPENDIX B4: Data of 40% Filler Loading



	Tensile strain at Break (Standard) (mm/mm)	Load at Break (Standard) (N)	Extension at Break (Standard) (mm)	Tensile extension at Break (Standard) (mm)
1	-1.07655	0.76735	-22.76838	-26.91386

	Time at Break (Standard) (sec)	Modulus (Automatic) (MPa)	Tensile stress at Total Creep (MPa)	Time at Yield (Offset 0.2 mm) (sec)
1	140.39201	99.81339	-----	-----

APPENDIX B5: Data of 50% Filler Loading