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Influence of TiO_2 on Mechanical, Microstructural, and Electrical Properties of TiO_2 /Rubber Composites

by

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A report submitted in fulfilment of the requirements for the degree of
Bachelor of Applied Science (Materials Technology) with Honours

**FACULTY OF EARTH SCIENCE
UNIVERSITI MALAYSIA KELANTAN**

2017

DECLARATION

I declare that this thesis entitled “Influence of TiO₂ on Mechanical, Microstructural, and Electrical Properties of TiO₂/Rubber Composites” is the result of my own research except as cited in the 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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Acknowledgement

I would like take this opportunity to express my deepest gratitude towards many people who have helped me directly or indirectly throughout the time doing this thesis.

First of all, I would like to mention and thank my supervisors Dr. Muhammad Azwadi Bin Sulaiman and Dr. Wannarat Panwiriyarat. They had shared a lot of knowledges, ideas, advices and helped me in many ways when I met a few problems during the course of experimental progress.

Next, I would like to give a thousand thanks to all my friends in UMK Jeli campus and PSU Surat Thani campus that had helped me and gave me tremendous supports. They were very willing to show and teach me the way to use some machines and softwares in the laboratories in order for me to work more efficiently.

Lastly, I would like to express my thanks to the staffs and laboratory assistants for always ready to assist, advise and help me in running some machines without delay.

Table of Contents

DECLARATION.....	i
Acknowledgement.....	ii
List of Figures.....	v
List of Tables.....	vi
List of Abbreviations.....	vii
Abstract.....	viii
Abstrak.....	ix
CHAPTER 1.....	1
INTRODUCTION.....	1
1.1 Background of the Study.....	1
1.2 Problem Statement.....	2
1.3 Research Objective.....	3
1.4 Expected Outcome.....	3
1.5 Significant of Study.....	4
CHAPTER 2.....	5
LITERATURE REVIEW.....	5
2.1 Dielectric Materials.....	5
2.2 Composite Dielectric Materials.....	5
2.3 Processing Methods.....	6
2.4 Characterisation Techniques for Flexible Dielectric Material.....	8
2.4.1 X-Ray Diffraction.....	8
2.4.2 Microstructure.....	9
2.4.3 Electrical Characterisation.....	10
CHAPTER 3.....	12
METHODOLOGY.....	12
3.1 Introduction.....	12
3.2 Raw Materials.....	14
3.2.1 Raw Material for Vulcanised Natural Rubber.....	14

3.3	Processing Method	14
3.3.1	Preparation of Vulcanised Natural Rubber	14
3.3.3	Compounding of Titanium Dioxide- Natural Rubber Composite	15
3.4	Characterization Techniques	15
3.4.1	X-Ray Diffraction (XRD)	15
3.4.2	Universal Tensile Testing Machine.....	15
3.4.3	Impedance Spectroscopy (IS).....	16
3.4.4	Rotorless Rheometer	16
Chapter 4	18
4.1	Raw Material Characterization.....	18
4.1.1	TiO ₂	18
4.1.2	Natural Rubber	19
4.2	Characterization of Different Loaded TiO ₂ Vulcanized STR.....	19
4.2.1	Curing Properties.....	19
4.2.2	Mechanical Properties	20
4.2.3	Electrical Properties	22
4.3	Characterization of Different Type of Rubbers.....	23
4.3.1	Curing Characteristics	23
4.3.2	Mechanical Properties	25
4.3.3	Electrical Properties	27
4.4	Morphological Properties.....	28
Chapter 5	30
Conclusion and Recommendations	30
5.1	Conclusion	30
5.2	Recommendations for Future Research	31
REFERENCE	32

List of Figures

No.	Title	Page
2.1	Role of Accelerators	6
2.2	Cross linkage Structures	7
2.3	XRD patterns of synthesized TiO ₂ powder	8
2.4	SEM micrographs BT/ENR50 composites	9
2.5	Computational results of static dielectric constant and static refractive index	10
3.1	Flow chart of research	12
3.2	Universal Tensile Testing Machine	14
3.3	Examples of datas obtained from a Rotorless Rheometer	15
4.1	XRD patterns of TiO ₂	16
4.2	Stress-Strain graph of STR 5L with varying TiO ₂ phr	18
4.3	Tensile strength and elongation at break of STR 5L compounds with varying TiO ₂ phr	19
4.4	Dielectric constant-Frequency graph of STR 5L with varying TiO ₂ phr	20
4.5	Rheograph of STR 5L, ENR 25 and ENR 50 compounds	21
4.6	Stress-Strain graph of STR , ENR 25 and ENR 50 compounds with 20 phr TiO	22
4.7	Stress-Strain graph of STR , ENR 25 and ENR 50 compounds with 20 phr TiO	23
4.8	Dielectric constant-Frequency graph of STR 5L, ENR 25 and ENR 50 composites with 20 phr	24
4.9	SEM micrographs of TiO ₂ /STR5L composites with various TiO ₂ loadings (a) 0, (b) 5, (c)10, (d) 20, (e) 30 and (f) 50 phr	25
4.10	SEM micrographs of TiO ₂ /ENR-25, ENR-50 composites with 20 phr (g) ENR-25 and (f) ENR-50	26

List of Tables

No.	Title	Page
3.1	Compounding formulation of vulcanized rubbers (STR, ENR 25 and ENR 50)	13
4.1	Cure characteristics of STR, ENR 25 and ENR 50 composites	17

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List of Abbreviations

TiO ₂	Titanium Dioxide
ZnO	Zinc Oxide
Zn	Zinc
XRD	X-Ray Diffraction
SEM	Scanning Electron Microscope
BT	Barium Titanate
STR	Standard Thai Rubber
ENR	Epoxidised Natural Rubber
MBT	Mercaptobenzothiazole

Abstract

In recent years, materials with high dielectric constant and tensile strength are highly sought after in various electronic applications and industries. Ceramic materials have some of the highest dielectric constant values but their applications are limited by their mechanical properties. On the other hand, elastomer exhibit high and beneficial mechanical properties but have low dielectric constant values. Hence, by producing composite from these two materials the limitations of each individual material will be solved. TiO₂ was chosen as filler to different types of rubber matrix, STR, ENR 25 and ENR 50, in this research due to its potential of high dielectric constant value and low cost. Tensile strength test on the STR composites with different TiO₂ loadings, 0, 5, 10, 20, 30 and 50 phr, show increase of stress at any point after 500% of strain from 1 MPa to 3 MPa but composite with 50 phr recorded lower values than composite with 30 phr due to the loading stress within the composite with 50 phr that affects the efficiency of network chains of rubber to orient themselves in the direction of stretching. ENR 25 and ENR 50 with 20 phr recorded 4.0 MPa and 4.1 MPa respectively which are significantly higher than STR at 500% of strain. Apart from that, the composites were also subjected to LCR test. At any point between 75 kHz to 30 MHz, the dielectric constant of the STR composites with different loadings increases as the filler loading increases within range of 2.9 to 3.4 dielectric constant at 10 MHz. In comparison between STR and ENR composites at 20 phr, ENR composites have significantly higher dielectric constant than STR composite where ENR 50 recorded the highest at 7.4.

Abstrak

Kebelakangan ini, bahan-bahan dengan pemalar dielektrik dan kekuatan tegangan yang tinggi sangat dicari dalam pelbagai aplikasi dan industry elektronik. Bahan seramik mempunyai antara nilai k yang tertinggi tetapi aplikasi mereka dalam industry terhad disebabkan kelemahan dalam kekuatan mekanikal. Sebaliknya, elastomer mempamerkan sifat-sifat mekanikal yang tinggi dan berfaedah tetapi mempunyai nilai k yang agak rendah. Oleh itu, dengan menghasilkan komposit dari kedua-dua bahan ini mampu mengatasi batasan yang ada pada individu bahan tersebut. TiO_2 telah dipilih sebagai pengisi dalam kajian ini kerana potensinya yang mempunyai nilai k yang tinggi dan kos rendah. Ujian Impedance Spectroscopy ke atas komposit2komposit yang telah dihasilkan dengan kandungan TiO_2 yang berbeza menunjukkan peningkatan nilai k dalam komposit-komposit tersebut. Selain itu, kekuatan tegangan bagi komposit telah diuji dan didapati pada kandungan TiO_2 yang tinggi kekuatan tegangan mula merosot disebabkan rangkaian getah tidak dapat mengorientasi diri mereka kea arah reganga



CHAPTER 1

INTRODUCTION

1.1 Background of the Study

In recent years, dielectric materials with high large electric energy storage are highly sought after in various electronic industries (Yang et al., 2008). Polymers have been popular for properties such as high tensile strength, low dielectric loss and low cost but is limited by their low dielectric constant. Ceramic materials on the other hand have some of the highest dielectric constant and large permittivity ceramics but have relatively low tensile strength in comparison to polymers. Hence, there are a lot of efforts in compounding polymers and high permittivity ceramic materials composites to combine mechanical properties and electrical properties of both the materials (Li et al., 2014).

TiO₂ is an electronic semi-conductor has free electrons that acts as current carriers. This is in contrast with the other type of semi-conductor which has “holes” as conduction (Earle, 1942). In modern days, titanium dioxide pigments are widely used in paints and plastics for exterior use (Day, 1990). Titanium dioxide is also found to be able to exhibit its dielectric property when used in a polymer-ceramic composite (Tchoul et al., 2010).

In 1791, William Gregor, an enthusiast of mineralogy in England has discovered the element Titanium. He studied some black sandy substance he found in his neighborhood containing mixture of iron oxide and a new metal which he

named menaccanite. Four years later, a German scientist named Martin H. Klaproth who was studying an ore that contain a never seen before chemical element from Hungary which he called titanium, after the Greek's mythology Titans. His research confirmed the discovery of William Gregor. However, Martin H. Klaproth was only able to successfully produce TiO_2 (Pappas, 2014).

Elastomer or natural rubber is a renewable polymer which exhibits exceptional properties such as high elasticity and durability that is suitable in a wide range of applications (Hamilton, 2016). Plantation rubber and rubber industry in Southeast Asia began when Wickham transferred Hevea, which provided over 30% of world's consumption in the early days, to the Orients. The discovery of vulcanization process by Goodyear and invention of better tapping methods by Ridley started a dramatic development in the rubber industry where Southeast Asia was one of top exporters until World War II (Hurley, 2006).

1.2 Problem Statement

Traditional polymer materials may exhibit high mechanical properties but has relatively low dielectric constant, less than 10 (Huang & Zhang, 2004). In order to increase the dielectric constant for better applications, polymer materials are used as matrix in composite with materials which has high dielectric constant as fillers (Zhang et al., 2002).

Compared to BaTiO_3 , TiO_2 has lower but still relatively high dielectric constant value ($k > 60$) among the binary metal oxides. However, it has a rigid anatase structure at low temperature (S. K. Kim et al., 2011). Hence, another material is needed to

provide the necessary mechanical properties. These needed properties can be found in natural rubber that has suitable mechanical and physical properties such as low temperature flexibility, high elasticity and fatigue resistance (González et al., 2016). However, it is important to identify the weight percentage of TiO_2 to be used as filler in natural rubber composite as high filler loading might decrease the mechanical properties of the natural rubber composite while low filler loading might affect the dielectric constant of the composite. The resulting product will be subjected to tests including dielectric constant and hardness.

1.3 Research Objective

The two main objectives in this study are:

- i) To produce TiO_2 /Rubber composites.
- ii) To compare properties between different types of rubbers as matrix.

1.4 Expected Outcome

The outcome of this research is expected to yield a flexible composite with the beneficial properties of TiO_2 with high dielectric constant. This outcome is expected as vulcanised natural rubber is highly chemically inert material which affirm the high dielectric constant of TiO_2 can be retained as no changes will occur to the chemical and microstructure of TiO_2 . Furthermore, natural rubber has been used in many studies

to be mixed with other high dielectric constant particles to produce similar expected composite.

1.5 Significant of Study

The purpose of this study is further improves and diversify the application of flexible dielectric constant composite. TiO_2 has suitable k value but its applications in electronic industries are retarded by mostly the mechanical properties of TiO_2 . Besides this, by improving the flexibility of TiO_2 based composites, more already in use applications can be improved and in some cases be used as an alternative to other materials with lower dielectric constant.

CHAPTER 2

LITERATURE REVIEW

2.1 Dielectric Materials

Dielectric materials are insulators. The simplest dielectric material is by inserting a layer of non-metallic solid between the plates of a capacitor. This material will increase its capacitance, acting as a none conducting bridge (Elert, 1998). When electric field is applied, an ideal dielectric material does not exhibit electrical conductivity. However, the material will abruptly conduct electricity when an applied electric field is above the critical field of the material. This happens as a cause of thermal instability due to the heat generated through conductivity or dielectric losses (David & Rethwisch, 2015)

2.2 Composite Dielectric Materials

According to Cava, (2000) the importance of dielectric ceramic materials is increasing due to the application in technologies especially microelectronics where the needs for high dielectric constant, low dielectric loss and low temperature dependence of dielectric constant is important.

Composite dielectric materials consist of two phase materials, conductor and an insulator. The need for properties that are not available in individual single

component materials drive the development of electrical composites. Hence, two or more components are combined to tailor composite materials to acquire the required properties (Almond & Rees, 2006).

2.3 Processing Methods

2.3.1 Vulcanization

Vulcanization is also known as crosslinking. This refers to the process of heating rubber, sulphur, and white lead causing crosslinking of the rubbers with substances such as sulphur. Accelerated vulcanization can complete the optimum curing in short periods compared to vulcanization without accelerators which could takes several hours. Typical accelerators consist of mixture of zinc oxide, ZnO and stearic acid. Figure 2.1 shows the role of accelerators such as ZnO in accelerating the process of vulcanization where Zn provides sites for the formation of complex compounds (Akiba, 1997).

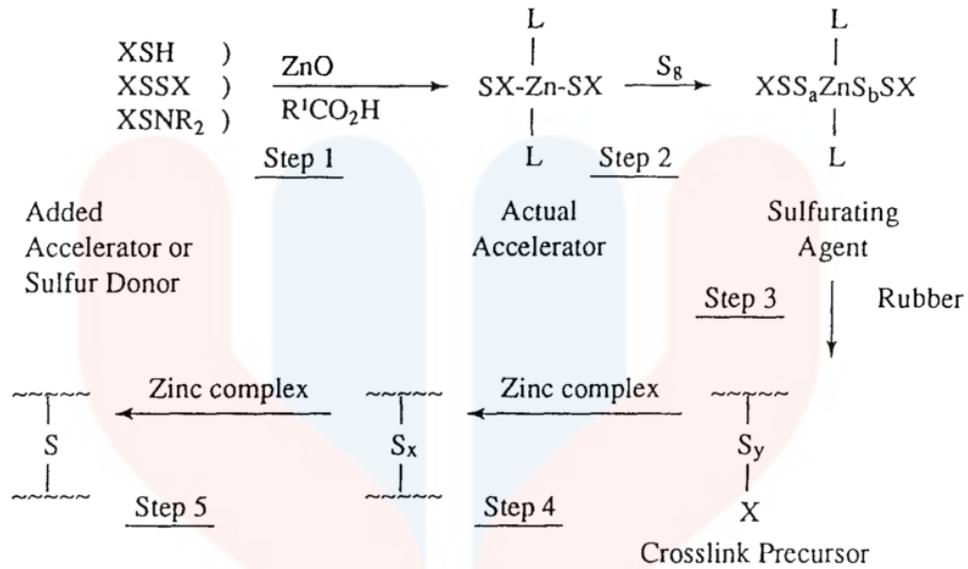


Figure 2.1: Role of Accelerators (Akiba, 1997)

Figure 2.2 shows the cross-link structure determined by the curing systems in the vulcanizates. Generally, there are mainly polysulfide (C-S_x-C) bonds, S donor forms C-S-C bonds while the peroxide system form C-C bonds (Guo *et al.*, 2016).

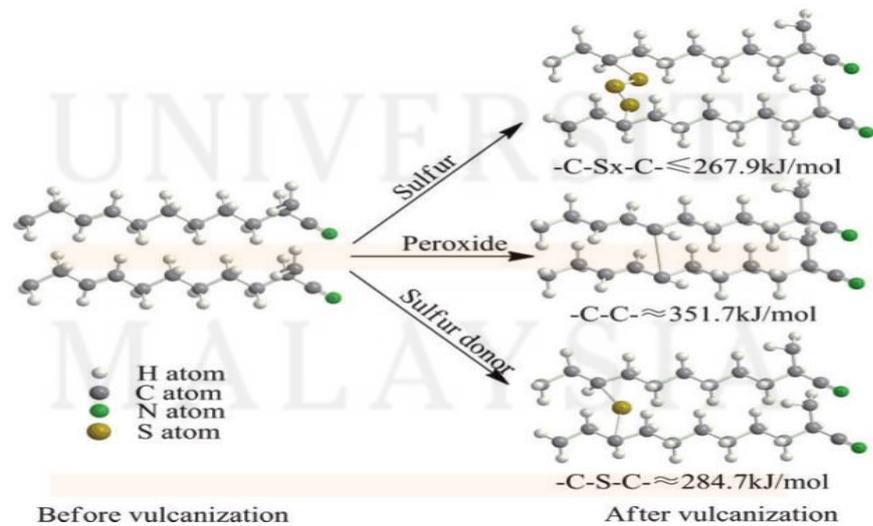


Figure 2.2: Cross linkage Structures (Guo *et al.*, 2016)

Vulcanization is usually carried out in an Internal Mixing Machine or Internal Mixer. An internal mixer has an enclosed chamber with two rotors inside. These rotors' function is to produce a kneading affect while the chamber ensures all materials which are added remain in place. The number of revolution of the rotors can be speeded up in order to reduce the time to mix a batch of materials to under 5 minutes (Min & White, 1985).

2.4 Characterisation Techniques for Flexible Dielectric Material

2.4.1 X-Ray Diffraction

X-ray powder diffraction (XRD) is a rapid analytical technique used for phase identification of a crystalline material and can provide information on unit cell dimensions. XRD is based on constructive interference of monochromatic X-rays and a crystalline sample that is finely grounded. The result is obtained by detect, process and count the diffracted X-rays produced by the constructive interference of incident rays when condition satisfy Bragg's Law. Then the sample is scanned through a range of angles to attain all possible diffraction directions. Identification of material is achieved by comparison of d-spacings with standard reference patterns (Dutrow & Clark, 2015). In Figure 2.3, two different forms of pure TiO_2 which are anatase and rutile exist in the synthesized TiO_2 powder sample (Kim et al., 2010).

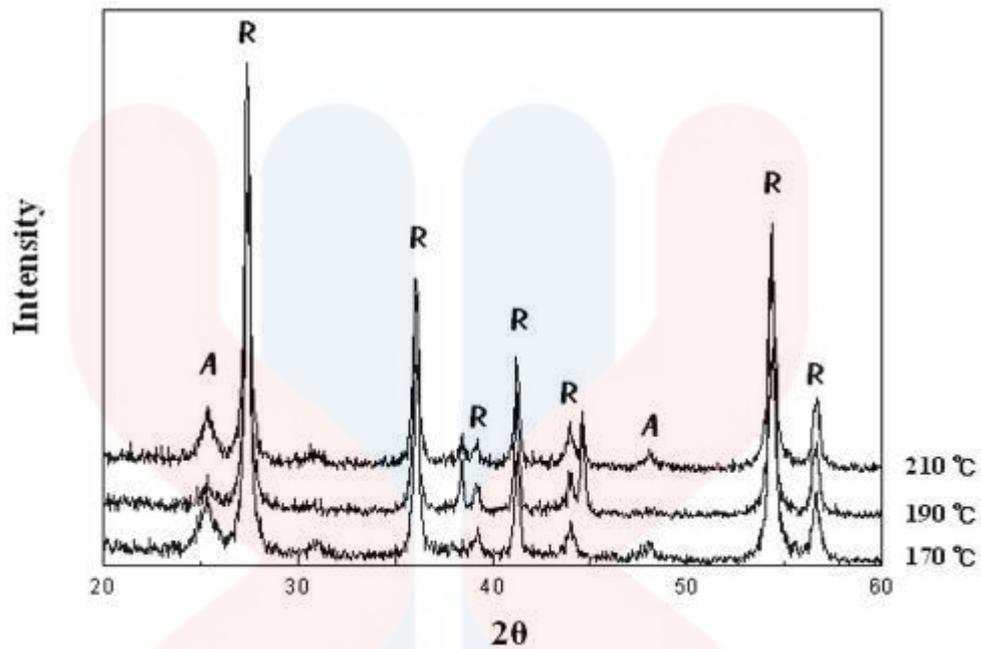


Figure 2.3: XRD patterns of synthesized TiO_2 powder (Kim *et al.*, 2010)

2.4.2 Microstructure

The microstructure of natural rubber- TiO_2 composite after been processed can be observed by using scanning electron microscope (SEM). SEM uses a beam of high energy electrons to reveal information about the sample such as external morphology, chemical composition, orientation of materials making up the sample and crystalline structure (Susan Swapp, 2015). By referring to a research involving a similar ceramic-polymer composite which uses natural rubber and BaTiO_3 , we can derive a hypothetical assumption on the dispersion of titanium dioxide in the natural rubber matrix. Figure 2.4 shows a SEM micrographs of BT/ENR50 composites with various BT loadings of 0, 5, 10, 15, 20, 30, 40 and 50 volume percentage represented by a, b, c, d, e, f, g and h respectively. At low particle loadings, the ceramic powder is uniformly dispersed in the ENR-50 matrix without apparent agglomeration. At higher loading

levels, BT starts to agglomerate due to the decreasing inter particle distance. At 40 and 50% loading levels, the composites become porous due to the poor adhesion between the matrix and ceramic particles (Salaeh, 2014).

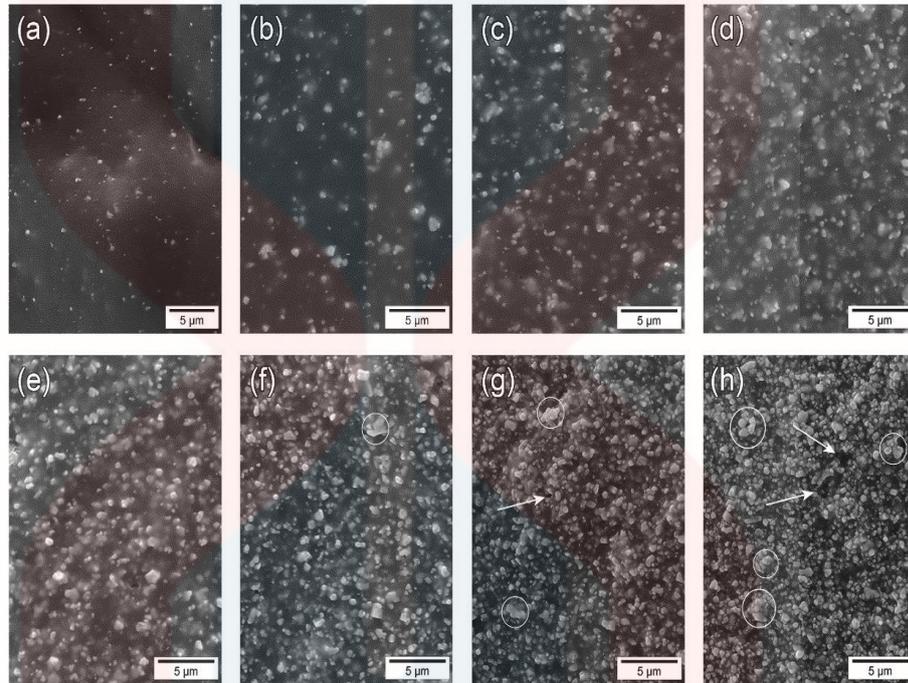


Figure 2.4: SEM micrographs BT/ENR50 composites (Salaeh, 2014)

2.4.3 Electrical Characterisation

Dielectric constant is also known as relative permittivity (Rouse, 2005). The most basic technique of measuring dielectric constant is the full sheet resonance (FSR). However, the accuracy limitations of full sheet resonance for measuring the dielectric constant of laminated materials due to irregular surface roughness (Deutsch *et al.*, 2002). Another way of testing for dielectric constant is split post dielectric resonator (SPDR). This technique can provide values for both x and y axis of a sample at a designated resonant frequency. The resonant frequency depends on both the sample's

permittivity and thickness (F. et al., 2016). Impedance spectroscopy is the easiest and most convenience technique to measure electrical properties. A wide range of frequency, typically 10^{-2} to 10^7 HZ is used to measure the impedance of a sample. Impedance must determine the resistivity and reactive components by alternating voltage across a sample and a standard resistor which are in series. The in and out phase components of the voltage are measured then divided by the magnitude of the current to acquire the resistivity and reactive components' impedance. In a ceramic sample, the different region characterized by resistance and capacitor are placed in parallel. After calculating the impedance spectrum, it is possible to identify the elements of resistance and capacitors which then can be quantified. In Figure 2.5 is an example of static dielectric constant and static refractive index of pure TiO_2 and doped TiO_2 by using Castep software. It shows an increase in dielectric constant as concentration of doping increases (Abed *et al.*, 2015)

Sample	static dielectric constant	static refractive index
TiO_2	6.14	2.47
Co/ TiO_2 15%	12.7	3.56
Co/ TiO_2 20%	27.9	5.29
Co/ TiO_2 25%	55.5	7.45

Figure 2.5: Computational results of static dielectric constant and static refractive index (Abed *et al.*, 2015)

CHAPTER 3

METHODOLOGY

3.1 Introduction

This study encompasses the compounding of polymer-ceramic composite where the natural rubber is used as matrix and titanium dioxide as the filler. The flow chart of this study is shown in Figure 3.1. A total of 8 samples will be produced. This process starts with the vulcanization of natural rubber and preparations of raw materials which include natural rubber, sulphur and accelerators such as zinc oxide, stearic acid and mercaptobenzothiazole. Then these materials are vulcanized at 160°C. the products are then subjected to moving die rheometer analysis. The compounding completes when the products are cured and hot pressed in compression moulding machine. The composites will be subjected to XRD, tensile strength test, LCR meter and SEM.

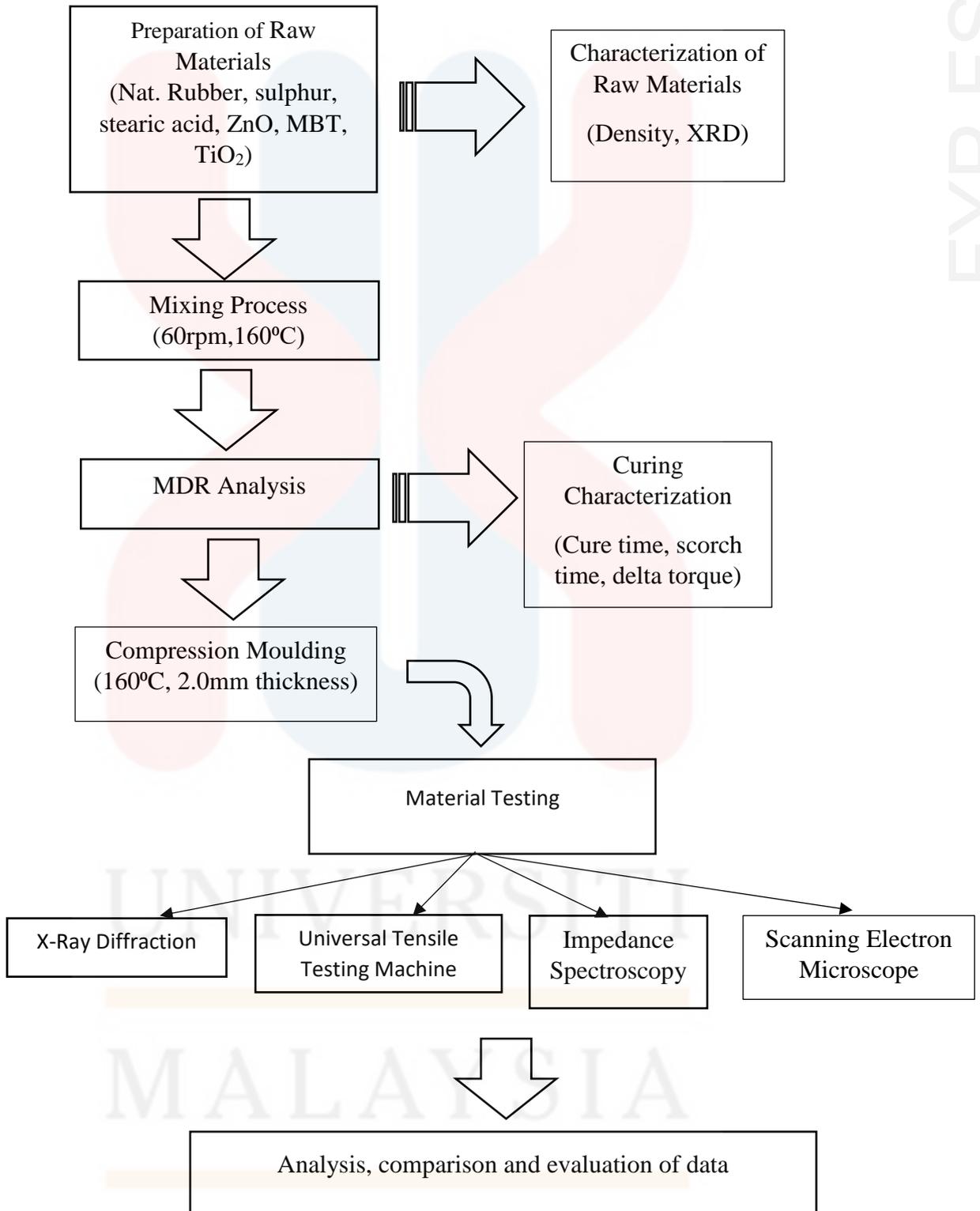


Figure 3.1: Flow chart of research

3.2 Raw Materials

3.2.1 Raw Material for Vulcanised Natural Rubber

The raw materials to prepare vulcanised natural rubber is rubbers which are Standard Thai Rubber (STR), 25% Expoxidised Natural Rubber (ENR 25) and 50% Epoxidised Natural Rubber (ENR 50), sulphur, zinc oxide (ZnO), stearic acid, and mercaptobenzothiazole (MBT). The density of the rubber will be determined.

3.3 Processing Method

3.3.1 Preparation of Vulcanised Natural Rubber

Natural rubber will be added with of sulphur of Zinc Oxide (ZnO), stearic acid, and mercaptobenzothiazole (MBT) of natural rubber. The mixture will then be heated at 160°C to maximise the vulcanization process (Arguello & Santos, 2016). As shown in Table 3.1, for every 100 g of natural rubber, 5.0 phr, 2.0 phr, 1.5 phr and 1.5 phr of ZnO, stearic acid, MBT and sulphur should be added in respectively (Salaeh, 2014).

Table 3.1: Compounding formulation of rubber (STR, ENR 25 and ENR 50)

Ingredients	Quantities
Rubber	100
ZnO	5.0
Stearic Acid	2.0
MBT	1.5
Sulphur	1.5

3.3.3 Compounding of Titanium Dioxide- Natural Rubber Composite

Vulcanised natural rubber will be mixed with 0, 5, 10, 20,30 and 50 volume percentage of TiO₂ powder in internal mixer at 160°C with rotor speed of 60rpm for 5 minutes. The materials are then collected and will be fabricated into thin form by using a compression moulding machine.

3.4 Characterization Techniques

3.4.1 X-Ray Diffraction (XRD)

Bruker D2-Phaser, Diffract. Eva software will be used for phase identification and morphology of TiO₂ powder. The scanning range will be used is between 20° to 90°. The X-ray diffraction pattern will be analysed by using DIFFRAC.EVA where the phase of the TiO₂ will be identified.

3.4.2 Universal Tensile Testing Machine

The universal tensile testing machine with an integrated ASTM can be used to identify the modulus of elasticity by analysing the stress-strain curves to understand the tensile property of the composites. The composites are first formed into thin sheets, then cut into 4 small dumb bell shaped samples of ASTM die type C. The grip separation speed was set to 500 mm/min and initial gauge length 20 mm following ASTM D412.



Figure 3.2: Universal Tensile Testing Machine

3.4.3 Impedance Spectroscopy (IS)

High frequency of IS ranging from 75kHz to 30MHz will be used to analyse the small composite blocks. At this frequency, the electrical properties of the composite such as dielectric constant, dielectric loss and resistivity will be attained.

3.4.4 Rotorless Rheometer

The Rotorless Rheometer is an apparatus used to measure the curing characteristics of elastomers such as delta torque, curing rate and scotch time (Albano et al., 2006). These informations can be used to determine the time the composites sit in the compression molding machine.



Figure 3.3: Examples of datas obtained from a Rotorless Rheometer

Chapter 4

RESULTS AND DISCUSSION

4.1 Raw Material Characterization

4.1.1 TiO₂

The TiO₂ used is commercialised product. The TiO₂ is examined by using XRD to produce patterns as shown in Figure 4.1 and obtained COD 7103589. The structure of TiO₂ peaked highest at 25.02° ,48.00° and 34.00° which correspond to hkl readings (-1 0 -1), (-2 0 0) and (-1 0 -5), which is the standard spectrum for anatase of TiO.

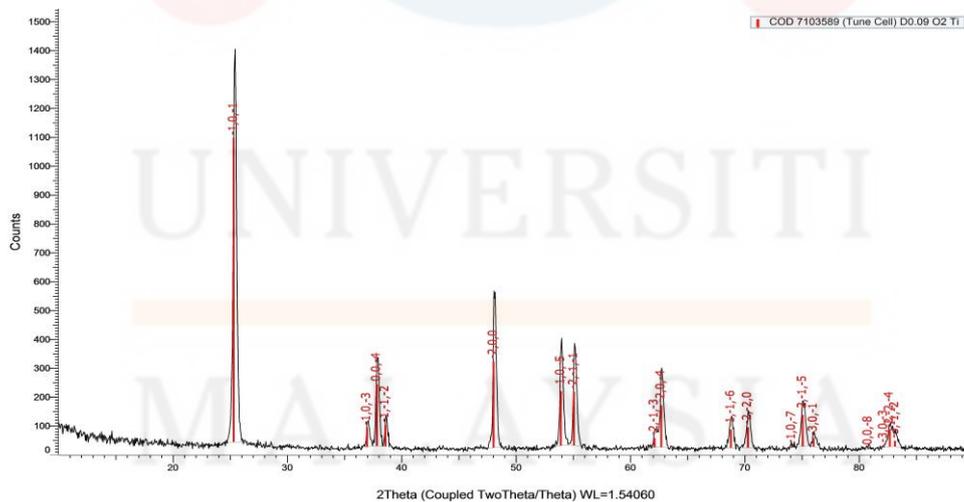


Figure 4.1 : XRD patterns of TiO₂

4.1.2 Natural Rubber

The density of natural rubber was weighed with an Archimedes principled weighing machine, the density obtained from the sample which was cut into $3 \times 3 \text{ cm}^3$ dimensions is 0.90 g/cm^3 , as per regular density of rubber (Jamil et al., 2006)

4.2 Characterization of Different Loaded TiO_2 Vulcanized STR

The STR rubber is first tested with different TiO_2 phr, 0, 5, 10, 20, 30 and 50 phr. Comparison among these composites will be made to identify the ideal TiO_2 content.

4.2.1 Curing Properties

Curing characteristic, cure time and torque were studied by using a moving die rheometer, MDRH 2020 model. From the Table 4.1 it can be seen the scotch time, t_{s2} , increase from phr loading of TiO_2 in STR 5L compounds from 0 to 10 phr 3.39 to 4.57 min before decreasing from 20 to 50 phr, from 4.52 to 3.51 min. The decrease may be due to lower content of natural rubber in the compounds hence lower vulcanization activities and increasing curing rate indexes are observed. The change of torque, delta torque increases along the increment of TiO_2 phr in STR 5L compounds. This shows the crosslink density increasing as the volume of STR 5L decrease.

Table 4.1: Cure characteristics of STR5L, ENR 25 and ENR 50 compounds

Testing at 160 ⁰ C x 30 min										
Compatibiliser	loading TiO ₂ (phr)	Cure characteristics								
		ML (dNm)	MH (dNm)	ts2 (min)	t90 (min)	MH-ML (dNm)	CRI (S ⁻¹)	HARDNESS	ts2	tc90
STR5L	0	1.04	8.54	3.39	7.52	7.50	0.40	24.4	219	472
	5	0.5	8.95	3.37	8.04	8.45	0.37	27.5	217	484
	10	0.58	9.68	4.57	9.17	9.10	0.38	27.4	297	557
	20	0.71	10.84	4.52	9.21	10.13	0.37	28.9	292	561
	30	0.86	11.60	4.25	8.16	10.74	0.43	30.7	265	496
	50	0.67	12.41	3.51	7.13	11.74	0.50	33.6	231	433
ENR25	20	0.43	13.83	1.05	2.03	13.40	1.72	34.4	65	123
ENR50	20	0.33	14.00	1.29	2.57	13.67	1.14	36.8	89	177

4.2.2 Mechanical Properties

The tensile strength of the compounds are tested with Tinius Olsen tensile test machine. The composites of STR 5L with TiO₂ phr loading of 0, 5, 10, 20 and 30 shows an increasing of stress at any point after 500% of strain but phr 50 recorded lower value than phr 30 as shown in Figure 4.2. This is due to the high loading of filler content affects the efficiency of network chains of rubber to orient themselves in the direction of stretching.

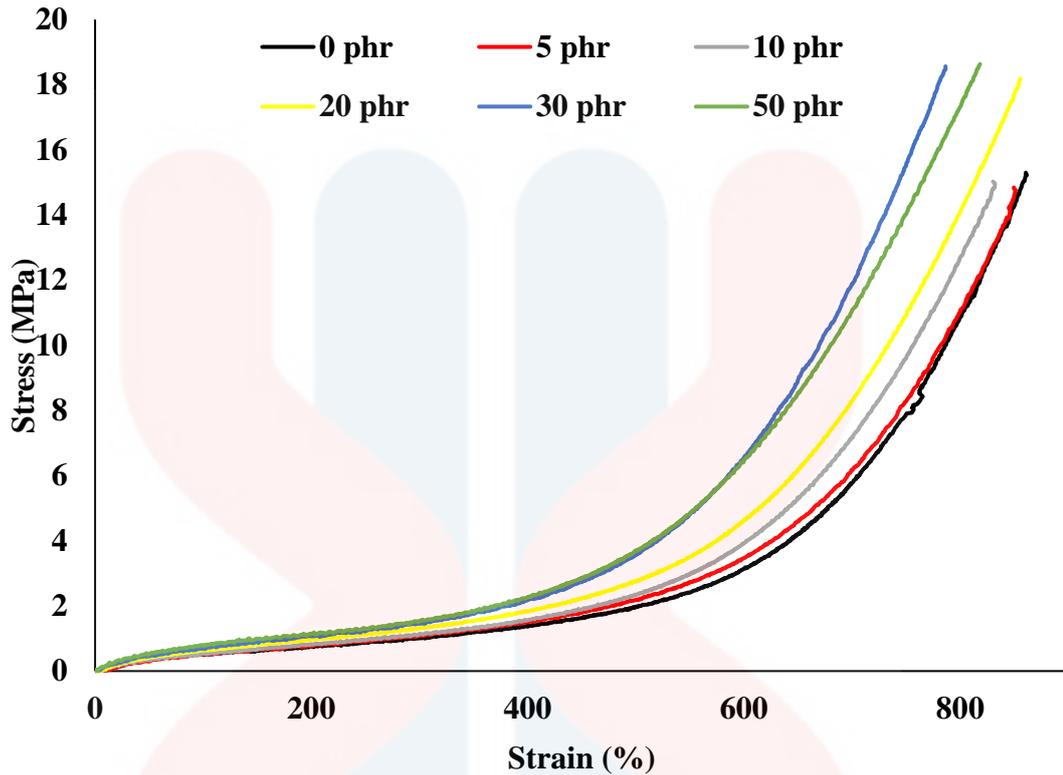


Figure 4.2: Stress-Strain graph of STR 5L with varying TiO₂ phr

Figure 4.3 shows the comparison of tensile strength and elongation at break of STR 5L compounds. The tensile strengths increase from phr 0 to 30, 14 MPa to 18 MPa, before decreasing at phr 50. This is due to the high filler loading which affects the tensile strength as explained before. Elongation at break is affected by the same reason as well. However, an irregular pattern was recorded for the elongation at break for the compounds of STR 5L may be caused by the irregular dispersion of TiO₂ in the compounds.

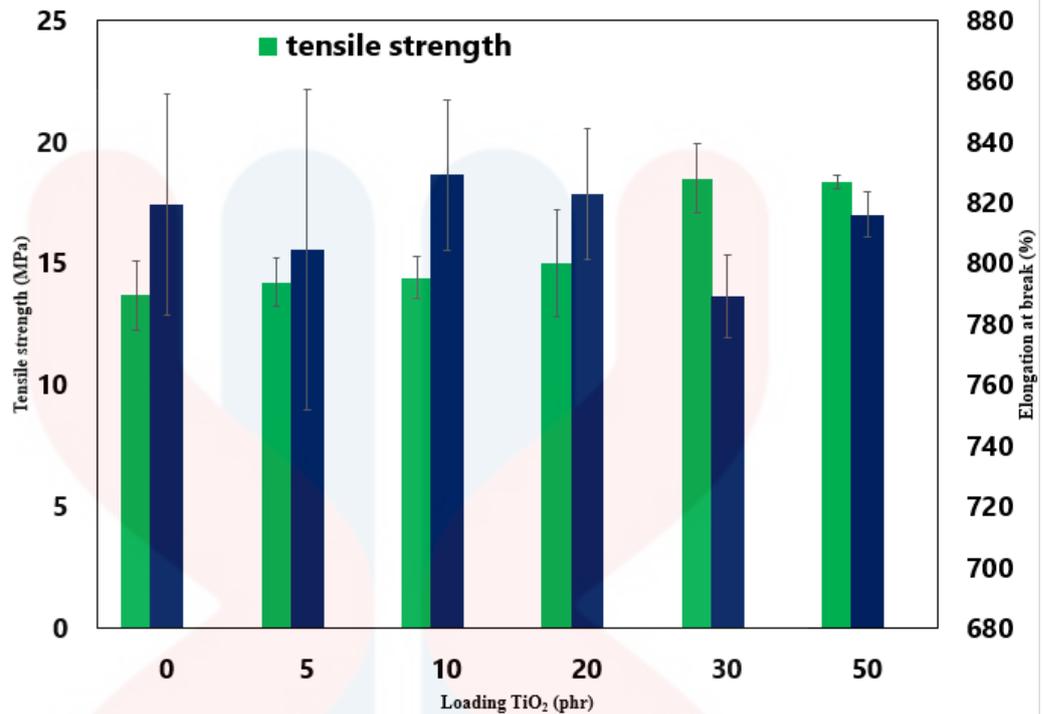


Figure 4.3: Tensile strength and elongation at break of STR 5L compounds with varying TiO₂ phr

4.2.3 Electrical Properties

The dielectric values of the composites were tested with LCR meter. Figure 4.9 shows dielectric constant-frequency graph of STR 5L with different filler loadings. The curves show a general decreasing dielectric constant values as frequency increases. The decrease happens as the dipole orientation and displacement polarization could not follow the change of alternation electric field efficiently at higher frequency. The increasing dielectric constant values in comparison among the STR composites are affected by the increase of TiO₂ content as shown in the Figure 4.4.

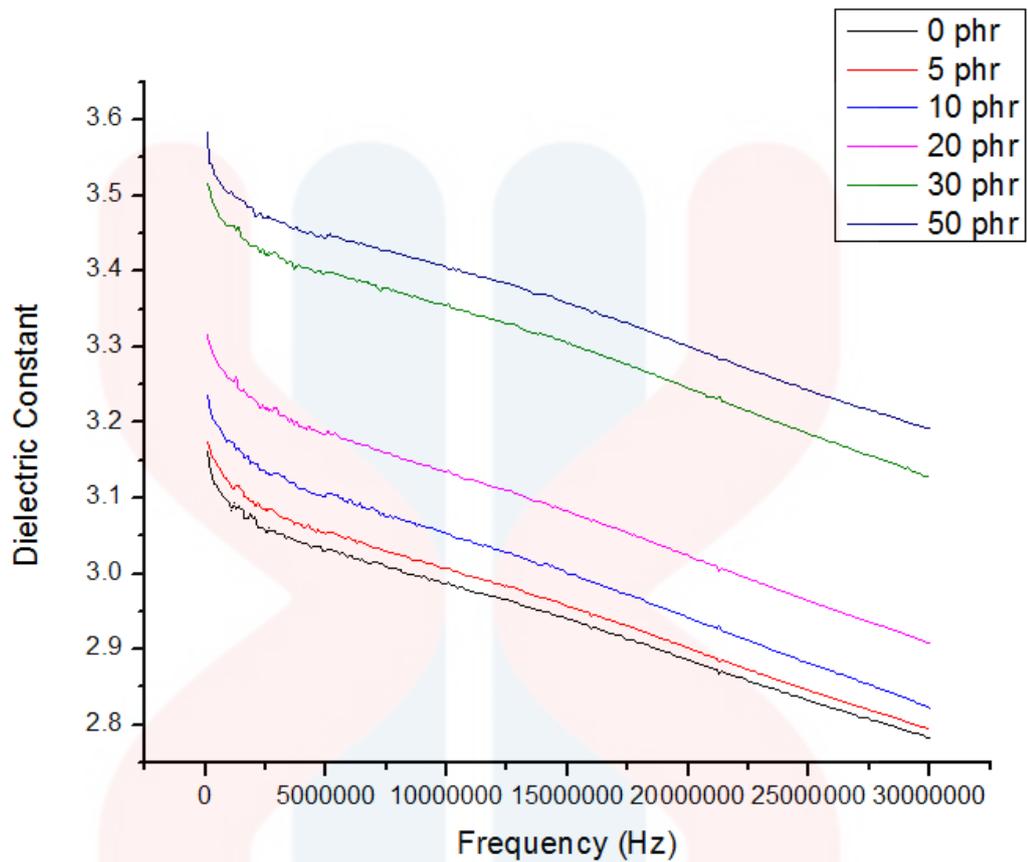


Figure 4.4: Dielectric constant-Frequency graph of STR 5L with varying TiO_2 phr

4.3 Characterization of Different Type of Rubbers

In comparison between STR, ENR 25 and ENR 50, 20 phr of TiO_2 was chosen as through observation and analysis of datas collected, STR composite with 20 phr shows the most promising potential of each characterizations made.

4.3.1 Curing Characteristics

. Figure 4.5 shows the comparison rheograph of 3 types of natural based rubbers: STR 5L, ENR 25 and ENR 50.

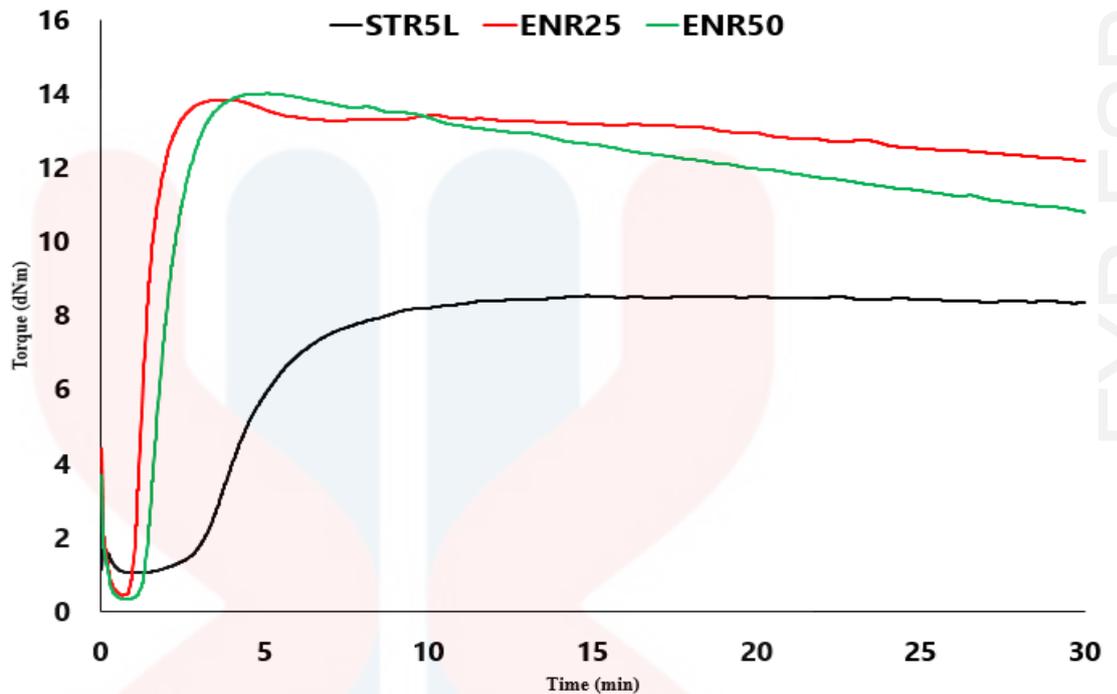


Figure 4.5: Rheograph of STR 5L, ENR 25 and ENR 50 compounds

The graph of STR 5L shows a plateau curve but reversion curves were observed for ENR 25 and ENR 50 where ENR 50 has a steeper curve. This shows the decreasing of torque as the testing time increase for the the ENR compounds. This can be attributed to the restructuring and changing structures of rubber vulcanizates after the maximum vulcanisation. Also, the breakdown of monosulfidic and ether linkages causes the reversion in the ENR networks. The ENR 50 compounds with higher percentage of epoxidation shows higher breakdown activities as the testing time increases. From Table 4.1, the ENR 25 and ENR 50 compounds showed shorter cure and scotch times but higher cure rate indexes due to the higher reactivity of the double bonds adjacent to the epoxide groups in the ENR molecules. ENR 50 compounds with more reactive double bonds takes longer scotch time than ENR 25 compounds. The ENR 25 and ENR 50 composites shows higher delta torque than STR composite due to the induced epoxide ring-opening reactions in the ENR compounds. Furthermore,

the delta torque increased with the increase of percentage of epoxidation of ENR compounds.

4.3.2 Mechanical Properties

. Figure 4.6 shows the stress-strain graph of STR, ENR 25 and ENR 50 compounds.

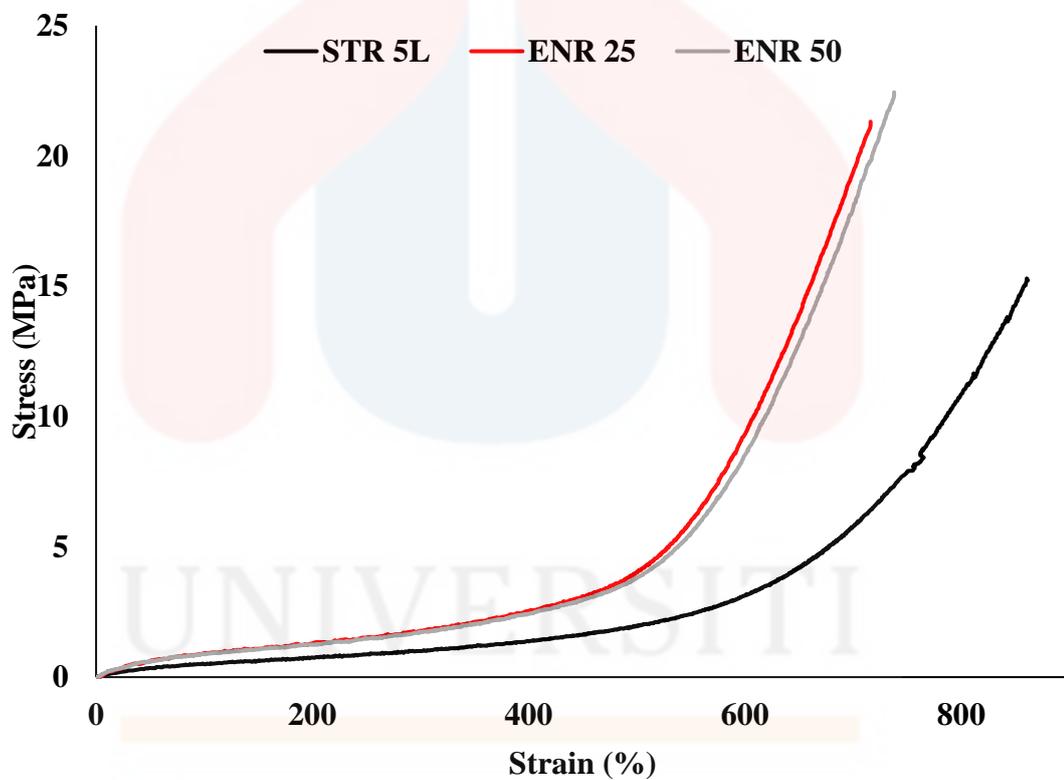


Figure 4.6: Stress-Strain graph of STR , ENR 25 and ENR 50 compounds with 20 phr TiO₂

The curves of all the compounds shows an increase in stress with increasing strain but a drastic increase is observed when strain is higher than 500%.

This occur as the result of “self-reinforcing” of the rubber caused by tendency of the

network chains of NR that tend to orient themselves in the direction of stretching or force applied leading to formation of crystallites. Compounds of ENR 25 and ENR 50 shows a significantly higher stress value at any point after 500% of strain than STR compounds due to the higher crosslink density of the compounds.

In comparisons with ENR 25 and ENR 50 as shown in Figure 4.7, the ENRs compounds has higher tensile strength, 19 MPa and 20 MPa respectively, due to both the stress-induced crystallization and high crosslink density. Furthermore, as shown in Table 4.1, the ENR compounds have higher hardness values than STR compounds due to the higher crosslink density too. The hardness of STR compounds with varying TiO₂ phr increases from 0 to 50 phr due to the addition of filler.

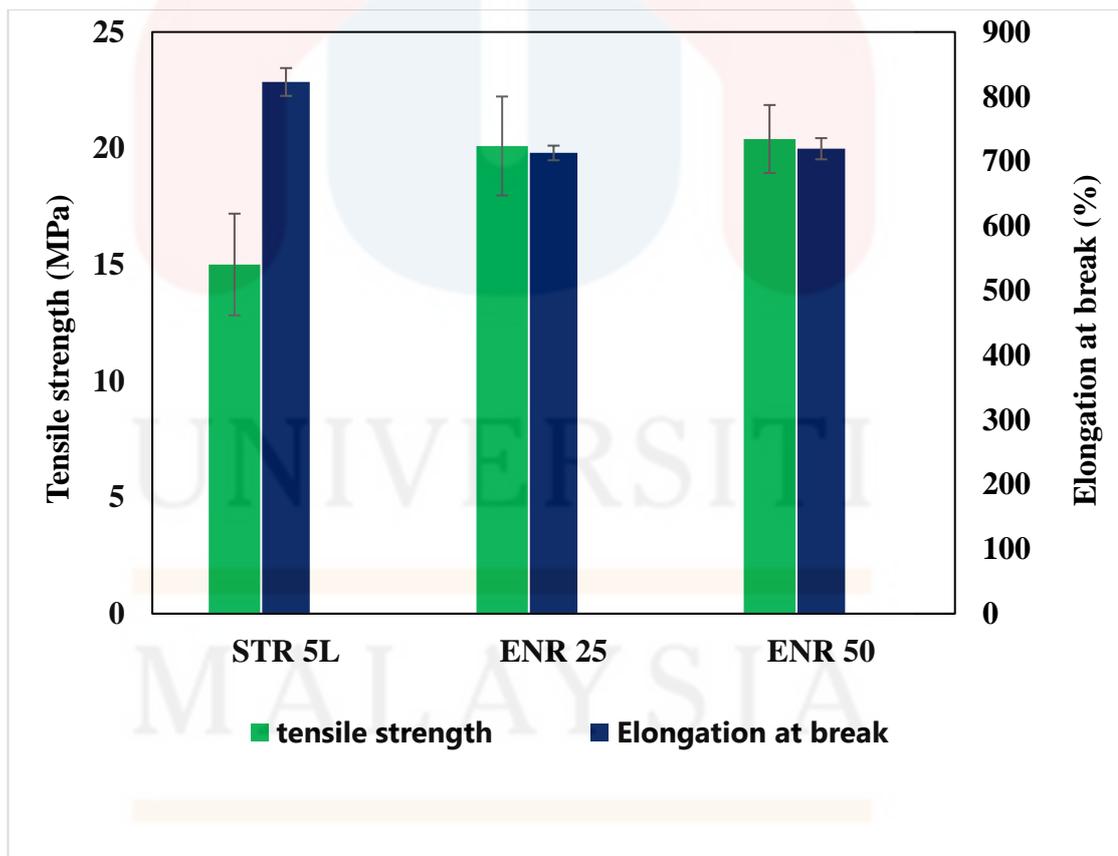


Figure 4.7: Bar chart of STR 5L, ENR 25 and ENR 50's tensile strength and elongation at break with 20phr of TiO₂

4.3.3 Electrical Properties

In Figure 4.8, it is shown that the composite of ENR 50 has the highest k value among the 3 different types of rubbers used to experiment while ENR 25 has second highest k values as frequency increases. This can be explained by the polarity of chains in the rubbers. STR 5L has no polar group in its structure unlike the ENR compounds. The ENR 50 which has higher percentage of epoxide groups than ENR 25 has higher polarity. High polarity in structure causes increase of dielectric constant of the rubber compounds.

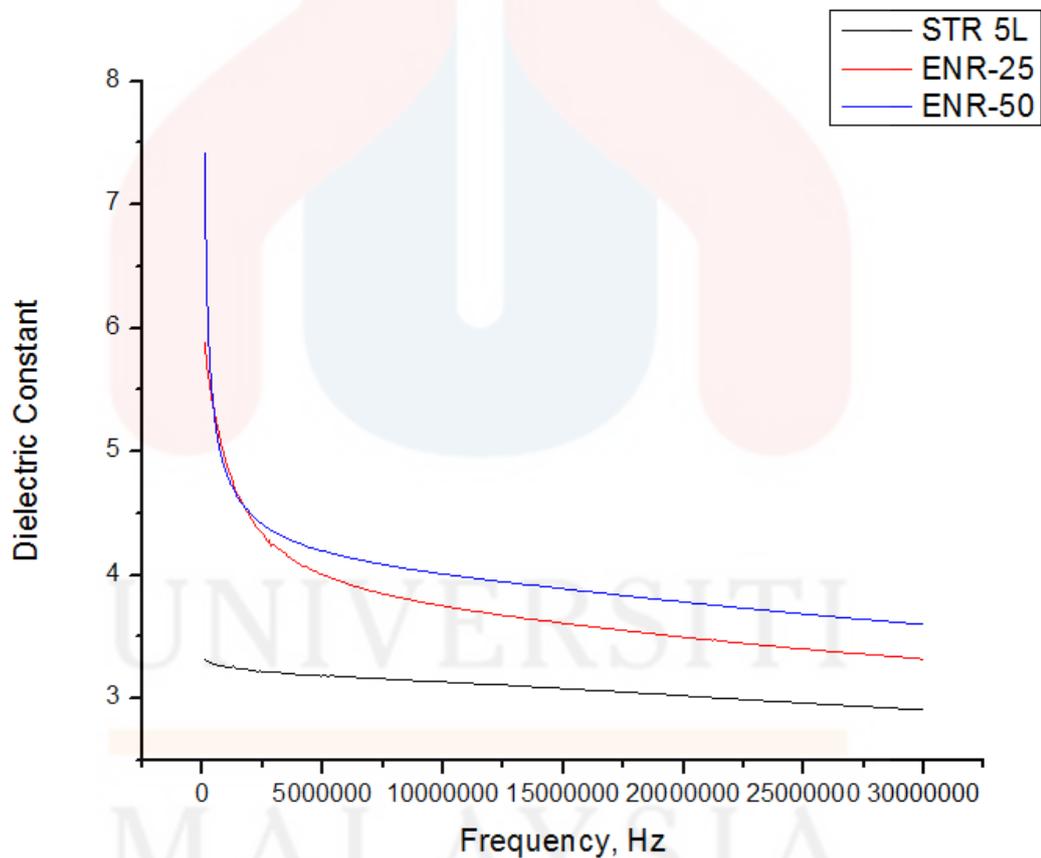


Figure 4.8: Dielectric constant-Frequency graph of STR 5L, ENR 25 and ENR 50 composites with 20 phr

4.4 Morphological Properties

The composites' morphologies are observed under the SEM to observe the dispersion of the fillers in the matrixes as shown in Figure 4.9 and Figure 4.10. Visible and homogenous morphologies can be observed with the exception of talcum powder particles as shown with blue arrows in (a) which was used to coat the composites to prevent them from sticking to the plastic sealants proves the mixing process used is appropriate. TiO₂ particles are fairly dispersed in (b) and (c) and still have high inter particle distance. However, from 20 phr onwards, (d), (e), (f), (g) and (h) larger speckles of TiO₂ start to exist. This is due to the decreasing inter particle distance which causes agglomeration of TiO₂ as shown by the red arrows in (d), (e) and (f). As the filler loading increases, the thickness of the matrix will decrease. The effect is apparent in (f) shown by the green arrow where porosity starts to occur at 50 phr. This happens as the adhesion between matrix and filler begins to decrease.

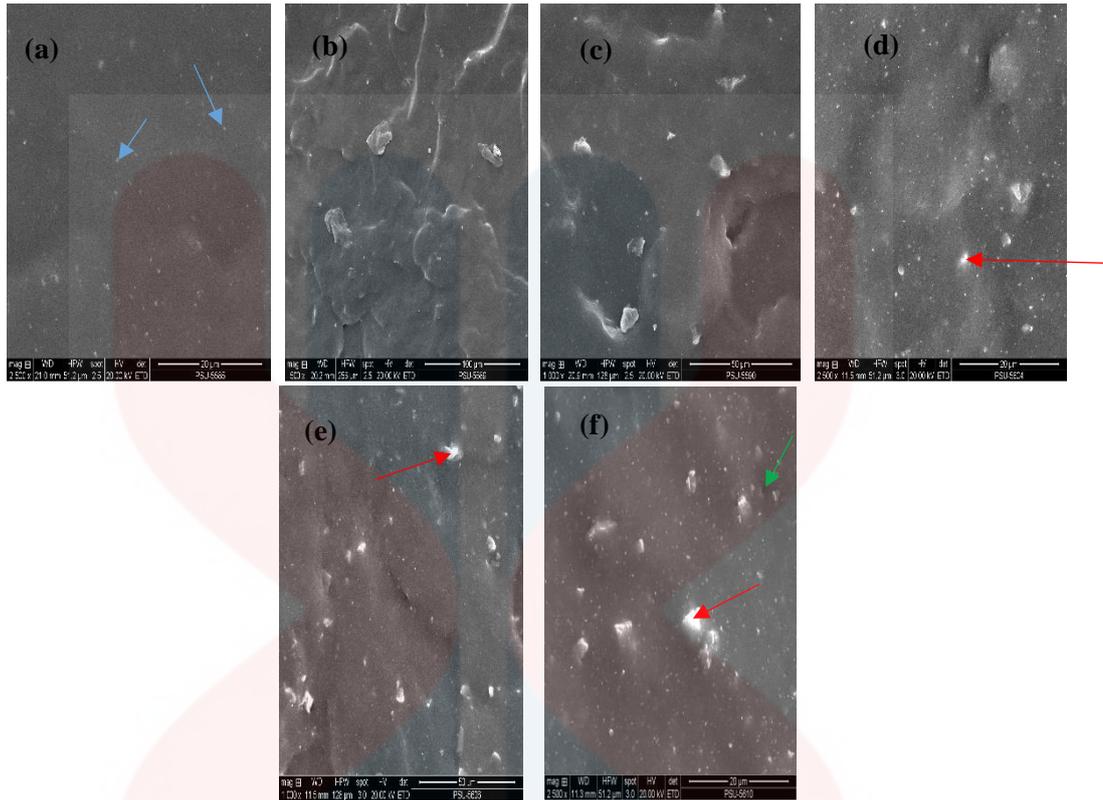


Figure 4.9: SEM micrographs of TiO₂/STR5L composites with various TiO₂ loadings (a) 0, (b) 5, (c) 10, (d) 20, (e) 30 and (f) 50 phr

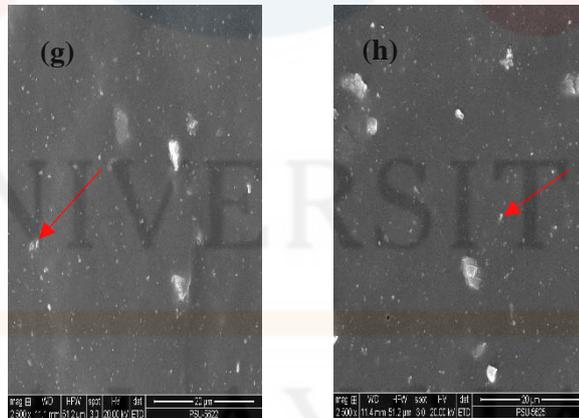


Figure 4.10: SEM micrographs of TiO₂/ENR-25, ENR-50 composites with 20 phr
(g) ENR-25 and (f) ENR-50

Chapter 5

CONCLUSION AND RECOMMENDATIONS

5.1 Conclusion

In this research, the raw materials that have been used were TiO₂ as filler and three different types of rubber, STR 5L, ENR 25 and ENR 50, as the matrix to form flexible dielectric composite. The manipulated variable in this research is the volume percentage of TiO₂ as filler in the composite (0, 5, 10, 20, 30 and 50 phr).

From the composites produced, a few analysis techniques, Impedance Spectroscopy, Tensile Test and SEM were used to identify to determine the best composition. Figure 4.6 shows that ENR 50 with 20 phr of TiO₂ have a more superior tensile strength among the three types of rubbers while STR 5L at 30 phr is the highest among the different loadings of STR 5L as shown in Figure 4.5. This shows that the TiO₂ content of the stated compounds have not affected the structure of the compounds. According to the observation on microstructure by SEM as shown in Figure 4.7, STR 5L with 20 phr shows good dispersion and adhesion to the matrix. Lastly, the dielectric constant of STR 5L with phr 50 and ENR 50 are the highest in their respective comparisons as shown in Figure 4.9 and Figure 4.10 respectively.

In conclusion, the composite STR 5L with 20 phr is the most suitable composition as compared with the others STR 5L with 20 phr shows fairly high dielectric constant, good dispersion of TiO₂ with no defects and unaffected tensile strength.

5.2 Recommendations for Future Research

There are recommendations that would like to be suggested for future research. First of all, dynamic mechanical analysis machine is used to analyse the compounds. This can help to further understand viscoelastic behaviour of the compounds at different temperature. Next, the dielectric constant of the compounds can be tested at different temperature to observe any drop in the values as reaction to the change of temperature.

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Appendice A



Figure A. 1: Sheeted Composite