

**STUDIES ON THE PROPERTIES OF CONDUCTIVE PARTICULATE
FILLERS FILLED RUBBER COMPOSITES**

by

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LIST OF SYMBOLS

M_H (dNm)	Maximum torque
M_L (dNm)	Minimum torque
t_2 (min)	Scorch time
t_{90} (min)	Cure time
R_v (Ω cm)	Volume resistance
E	elastic modulus
μ	micron (10^{-6})

LIST OF ABBREVIATION

ABS	Acrylonitrile butadiene styrene
ACA	Anisotropic conductive adhesive
ASTM	American Standard for Testing and Materials
CB	Carbon black
CBS	N-cyclohexyl-2-benzothiazole sulfonamide
CF	Carbon fibre
CRF	Channel replacement furnace
ECA	Electrically conductive adhesive
ECF	Extra conductive furnace
EMI	Electromagnetic interference
EMIS	Electromagnetic interference shielding
ENB	5-ethylidene-2-norbornene
EPDM	Ethylene-propylene-diene rubber
ESD	Electrostatic discharge
EVA	Ethylene vinyl acetate
G	Graphite
GPF	General purpose furnace black
HAF	High abrasion furnace black
HDPE	High Density Polyethylene
ICA	Isotropic conductive adhesive
IPPD	N'-phenyl-p-phenylene diamine
ISAF	Intermediate Super Abrasion Furnace Black
LDPE	Light Density Polyethylene
NBR	Nitrile rubber
NR	Natural rubber
phr	Part per hundred rubber
RFI	Radiofrequency interference

RRIM	Rubber Research Institute of Malaysia
SCF	Super Conductive Furnace
SEM	Scanning Electron Microscopy
SMR	Standard Malaysian Rubber
TGA	Thermo gravimetric Analysis

STUDIES ON THE PROPERTIES OF CONDUCTIVE PARTICULATE FILLERS FILLED RUBBER COMPOSITES

ABSTRACT

The use of conductive fillers on the properties of rubber composites was studied. Three types of conductive filler have been used in this study, i.e. aluminium, zinc and carbon black. Natural rubber (NR) and Ethylene-Propylene-Diene terpolymer (EPDM) were chosen as a matrix. Compounding was carried out using a two-roll mill based on a semi-efficient vulcanization (semi-EV) system. The investigation of rubber composite properties involves curing characteristics, mechanical and electrical properties, thermogravimetric analysis (TGA), morphological study and swelling behavior. The scorch and cure time of aluminium and zinc filled natural rubber composite decreased while maximum torque increases with increasing filler loading. Zinc filled natural rubber composites exhibit higher tensile strength and elongation at break but lower tensile modulus compared to aluminium filled natural rubber composites. Zinc filled natural rubber composites show higher crosslink density than aluminium filled natural rubber composites. However, the addition of aluminium and zinc filler reduced the electrical resistivity and increased the thermal stability of the natural rubber. The presence of aluminium fillers in EPDM composites shortens the scorch and cure time. Aluminium filled natural rubber composites exhibit higher tensile strength, tensile modulus and elongation at break compared to aluminium filled EPDM composites. Thermogravimetric analysis (TGA) study shows an increment in thermal stability after the addition of aluminium filler in natural rubber and EPDM rubber composites. On the other hand, EPDM rubber composites show higher thermal stability and crosslink density compared to that of natural rubber composites. The presence of carbon black in the Al/CB system increased the thermal stability of Al/CB hybrid composites. Tensile

strength and tensile modulus increased with increasing carbon black in the Al/CB hybrid NR system. The electrical resistivity of natural rubber composites decreased with the addition of carbon black in the system. In addition, the swelling behavior study shows that the crosslink density can be increased by replacement of aluminium with carbon black due to the increment in rubber-filler interaction. Thermal ageing of rubber composite at 100 °C for 48 hours decreased the tensile strength due to the deterioration of rubber matrix.

KAJIAN KE ATAS SIFAT-SIFAT KOMPOSIT GETAH TERISI PENGISI- PENGISI PARTIKEL KONDUKTIF

ABSTRAK

Penggunaan pengisi-pengisi konduktif ke atas sifat-sifat komposit getah telah dikaji. Tiga jenis pengisi konduktif telah digunakan dalam kajian ini iaitu aluminium, zink dan hitam karbon. Getah asli dan Ethylene-Propylene-Diene terpolymer (EPDM) telah dipilih sebagai matriks. Penyebatian telah dijalankan menggunakan penggiling bergulung dua berdasarkan sistem pemvulkanan separuh cekap (semi EV). Penyelidikan sifat-sifat bahan komposit getah melibatkan ciri-ciri pematangan, sifat-sifat mekanikal dan elektrik, analisis termogravimetri (TGA), kajian morfologi dan tingkah laku pembengkakan. Masa skorj dan pematangan komposit getah asli terisi aluminium dan zink menurun manakala tork maksimum meningkat dengan peningkatan beban pengisi. Komposit getah asli terisi zink mempamerkan kekuatan tensil dan pemanjangan pada takat putus yang lebih tinggi tetapi modulus tensil yang lebih rendah berbanding dengan komposit getah asli terisi aluminium. Komposit getah asli terisi zink menunjukkan ketumpatan sambung silang yang lebih tinggi daripada komposit getah asli terisi aluminium. Walaubagaimanapun, penambahan pengisi aluminium dan zink telah menurunkan kerintangan elektrik dan meningkatkan kestabilan terma getah asli. Kehadiran pengisi-pengisi aluminium di dalam komposit getah EPDM telah memendekkan masa skorj dan pematangan. Komposit aluminium terisi getah asli telah mempamerkan kekuatan tensil, modulus tensil dan pemanjangan pada takat putus yang lebih tinggi jika dibandingkan dengan komposit aluminium terisi EPDM. Kajian analisis termogravimetri (TGA) menunjukkan satu peningkatan dalam kestabilan terma selepas penambahan pengisi aluminium dalam komposit getah asli dan getah EPDM. Selain itu, komposit getah EPDM menunjukkan kestabilan terma dan ketumpatan sambung silang yang lebih tinggi berbanding dengan komposit getah asli.

Kehadiran hitam karbon dalam sistem Al/CB juga meningkatkan kestabilan terma komposit getah asli hibrid Al/CB. Kekuatan tensil dan modulus tensil meningkat dengan meningkatnya hitam karbon dalam sistem Al/CB. Kerintangan elektrik bagi komposit getah asli menurun dengan penambahan hitam karbon dalam sistem tersebut. Sebagai tambahan, kajian tingkah laku pembengkakan menunjukkan bahawa ketumpatan sambung silang boleh ditingkatkan dengan penggantian aluminium oleh hitam karbon disebabkan oleh peningkatan dalam interaksi getah-pengisi. Penuaan terma komposit getah pada suhu 100 °C selama 48 jam menyebabkan kekuatan tensil menurun disebabkan oleh kemerosotan matriks getah.

CHAPTER 1 INTRODUCTION

1.1. Overview of conductive rubber composites

Studies on conductive rubbers are nothing new (Sau et al., 1998a; Das et al., 2002; Wan et al., 2005). Conductive rubber has been known since the latter part of nineteenth century, but it was not major interest until about 1930, when several patents described the use of conductive composites for the prevention of corona discharges in cables (Norman, 1970). The use of conductive rubber for heating purposes has been the subject of numerous patents since about 1940, but there are considerable difficulties associated with the inherent variability and the liability of materials to change in resistance on flexing. Conductive silicone rubbers show some promise in overcoming this difficulty. Generally, rubbers having resistances below 10^4 ohms are considered conductive; between 10^4 and 10^8 ohms, rubbers are considered antistatic; and between 10^8 and 10^{16} ohms, rubbers are considered insulating (Nagdi, 1993; Jiang et al., 2005).

One of practical methods to obtain conducting rubber composites is adding conductive fillers like carbon blacks, carbon fibers, metallic powders, flakes and fibers as well as metallic hydrides into rubber matrices (Das et al., 2002; Wan et al., 2005). In general, the resistivity of common metal fillers such as silver and copper are 1.59×10^{-6} ohm cm and 1.7×10^{-6} ohm cm, respectively (Bhattacharya, 1986). Furthermore, the resistivity of carbon black is 1.5×10^{-3} ohm cm (Sau et al., 1998a). From the resistivity value, it is noted that metal fillers exhibit lower resistivity compared to carbon black. Conductive rubber composites have been developed for numerous applications, such as electromagnetic interference shielding, self-regulating low-temperature heater, self-resetting over-current protection elements, electrostatic charge dissipation and many different sensors for vapors, chemicals, mechanical stress and temperature.

It was observed experimentally that the electrical conductivity of conductive particulate filler-rubber composite predominantly depends on the electrical conductivity of conductive fillers, the particle shape and size, and the volume fraction and the spatial arrangement of the particles (Bhattacharya, 1986; Krupa et al., 2004; Xue, 2004). In many applications high conductivity is main requirement and the mechanical properties of such a system may be a secondary consideration.

Various rubbers are used for preparation of such composites, for example natural rubber, silicone rubber, nitrile rubber (NBR), butyl rubber, and ethylene-propylene-diene terpolymer (EPDM). Even though rubbers are normally good insulators with relatively high electrical resistivity, the nonpolar rubbers being better resistivity than the polar ones, which means the polar rubbers shows better electrical properties than nonpolar rubbers (Nagdi, 1993; Sau et al., 1998a). The electrical properties of rubber composites are more dependent on the compounding ingredients than on the basic rubber. During conductive fillers filled rubber compounding, there are few parameters need to be considered such as types of conductive fillers and types of matrix used in the composites systems. Hence, these two parameters were considered in the present research project.

Most of the time, carbon blacks are chosen as fillers for conducting rubbers and conducting plastics because of their low cost, low density, high electrical conductivity and in particular, specific structures that enable a formation of conductive network inside a polymer matrix at relatively low filler concentration. Judging from low resistivity of metal filler compared to carbon black, the partial or completely replacement of metal fillers by carbon black in rubber matrix need to be justified. However, it is realized that the use of metal filled rubber composites should be optimized in order to challenge the achievement of carbon black in rubber which are widely used in the current applications.

1.2. Objectives of the study

This study aims to give better understanding on electrical, mechanical as well as thermal properties of conductive rubber composites especially on the metal filled rubber conductive composites. The present work is conducted based on the following objectives:

- i. To study the effect of aluminium and zinc filler on the properties of natural rubber (SMR L) composites.
- ii. To study the effect of difference matrices i.e. natural rubber (SMR L) and ethylene-propylene-diene terpolymer (EPDM) on the properties of aluminium filled composites.
- iii. To elucidate the effect of partial or completely replacement of aluminium fillers by carbon black on the properties of natural rubber (SMR L) composites.

CHAPTER 2 LITERATURE REVIEW

2.1. Introduction

There are four ways to make plastics conductive. The first route is conductive polymers. The charge transport in conductive polymer is believed to occur through a combination of two primary mechanisms, propagation of charge along the polymer chain and hopping of charge between neighbouring chains (Margolis, 1989; Gal et al., 2006). For example polyacetylene, a conjugated organic polymer could attain high level of electronic conductivity when oxidized by suitable reagents (Campbell, 2001; Springborg, 2002; Gal et al., 2007). The concept of conductivity and electroactivity of conjugated polymers was quickly broadened from polyacetylene to include a number of conjugated hydrocarbon and aromatic heterocyclic polymers, such as polyphenylene. These polyacetylene and polyphenylene based compound have proved to be candidate materials for conductivity, but problem still remain with processability, mechanical properties, and stability.

The terms ionic conducting polymers is used when referring to polymer system that have a high ionic conductivity. The ability of a polymer to conduct an ion or ions may be either desirable or disastrous. It is not surprising that fast ionic conduction in polymers is currently an area of great interest. This interest is to understand the ionic conduction mechanism in polymers and to use these polymers applications such as high energy density batteries, electrochromic displays, specific-ion sensors, and other electrochemical devices that capitalize on the unique electronic, ionic, and mechanical properties of ionic conducting polymers.

The third approach is to apply a metallic plating and coating on the moulded plastic parts, a two step process which is becoming obsolete because of the cost

involved and the difficulties in obtaining a good coating (Taguchi et al., 2006). Metallic plated and coated plastic products are used primarily for decorative applications, automotive and appliance products, plumbing and marine applications, and other consumer and industrial products. Conductive plastic products by metallic plating and coating are used to provide electrostatic discharge (ESD) and electromagnetic interference-radiofrequency interference (EMI/RFI) shielding. Most metallized plastics products are made by electroplating, with the use of electroless plating increasing for EMI/RFI applications. The other principal processes are vacuum metallizing, sputtercoating (a vacuum process), arc spray and flame spray, and hot foil stamping.

The last option is to incorporate conductive fillers in the polymer matrix. Metal, carbon, or metal-coated fillers in the form of particles and fibers are widely used as conductive fillers (Wu et al., 2007). Metal fillers have a higher conductivity among the conductive fillers. Metal filled plastics have emerged as a new material with potential as an alternative to metal in specific applications. This potential derives primarily from the substantial modification of the thermal and electrical characteristics of plastics when combined with metallic fillers. In addition, metal filled plastics retain some of the advantage of plastics (e.g., cost, fabrication, maintenance, density, and aesthetics).

2.2. Type of conductive fillers

2.2.1. Introduction

Generally, fillers are used in either fibre or particulate form. Additions of fibres into polymers have a tendency to improve stiffness, strength, hardness, abrasion resistance, heat deflection temperature, and lubricity whereas reducing shrinkage and creep. The fibre aspect ratio (that is, the ratio of the length L to the diameter D of a fibre) and the orientation of fibres (that is, whether the fibres are globally oriented in one direction or laminated in alternating layers, plies, or mats) can have a profound

influence on the properties of the composites (Boo et al., 2007). Commonly, the most effective reinforcing fillers are fibres which intrinsically have a high elastic modulus and tensile strength.

Unlike fibres, particulate fillers have low aspect ratios, often approximating those of flake, sphere or plates. None of the particulate fillers improves the ultimate tensile strength of thermoplastics. Actually, tensile, flexural, and impact strength are normally lowered, in particular at higher filler contents. While hardness, heat deflection temperature, and surface finish may be enhancing, the property which is mainly improved is stiffness. Thermal expansion, mould shrinkage, extensibility, creep are reduced.

2.2.2. Metal Fillers

Since metal are the quintessential conductors of electricity, it is not surprising that they have been used as fillers to create conductive polymer composites (Das et al., 2002; Boudennea et al., 2005; Wan et al., 2005). In comparison to conductive carbon, the resistivity of metals are much lower, while their densities are higher. Table 2.1 shows the properties of common metal fillers. Only aluminium has a density comparable to carbon. Until recently, the most common form of metal fillers was either spherical or irregularly shaped isotropic particles. These particles were typically 100 μm or less in diameter. These small isotropic particles are needed in high concentrations to produce an electrically conductive composite. Mechanical properties and density advantages are usually lost because of the high filler loadings required (Bhattacharya, 1986). In the past few years, metal fibres, flakes, and ribbons have become available. In these anisotropic forms metals are more attractive as fillers because reduced concentration levels are possible.

Several factors influence the use of metals in polymers. Copper, for example, catalyzes the degradation of many polymers (Bhattacharya, 1986). Iron, copper, and aluminium oxidize rapidly at the particle's surface. These oxide layers are non-conductors and therefore dramatically change the electrical properties of the composite. This effect is most significant with smallest particles, because the oxide layers comprise a greater fraction of particle than they do on larger particles. For example, the resistivity of oxidized aluminium particles may be as high as 10^5 ohm-cm. The oxide layer on aluminium particles will effectively insulate the core, making composites nonconductive up to very high filler concentrations. In both cases, the particles used were under 100 μm in size. Since all metals except gold oxidize, the long-term electrical stability of metal plastic composites must be investigated. Of the repeatedly used metals, silver is the most conductive. It is also the most expensive. Stainless steel is very resistant to oxidation, although it is less conductive than other metals (it is still more conductive than carbon-base conductor) and more dense. Aluminium is attractive because it is a low-cost, low-density, highly conductive metal. Surface oxidation is the primary electrical concern when using aluminium. Nickel is also of interest because of its low rate of oxidation.

2.2.3. Carbon fillers

Among different type of conductive fillers, carbon filler is the most widely used materials for polymer matrices (Sau et al., 1998a). Carbon fillers are regularly employed in the production of polymer composites to enhance electrical conductivity and mechanical properties, as well as to maintain light weight and corrosion resistance (Sau et al., 1998a; Thongruang et al., 2002). Conductive carbon fillers of current commercial relevance include carbon black, graphite, and carbon fibre, which differs greatly in both structure and form.

Table 2.1: Summary of properties of metal fillers used to produce conductive composites (Bhattacharya, 1986)

Metal	Density (g/cm ³)	Resistivity (ohm-cm)	Size	Form
Copper	8.82	1.7×10^{-6}	~44 μm	Dendritic
			~100 μm	Dendritic
			~58 μm	Flake
			<50 μm	Electrolyte
			<100 μm	Spherical
Nickel	8.90	6.84×10^{-6}	52-74 μm	Irregular
			~4 μm	Spherical
			10-30 μm	Irregular
			~20 μm	Irregular
			0.001mm x 2 x 2 μm	Flake
Aluminium	2.71	2.8×10^{-6}	~5 μm	Irregular
			~100 μm	Irregular
			<100 μm	Spherical
			0.1 mm x 1.25 μm	Fibres
			0.25mm x 1 x 1.25 μm	Flakes
Iron	7.86	10×10^{-6}	25-100 μm	Irregular
Stainless steel	7.80	74×10^{-6}	4-50 μm diam.	Fibres
			4-79 μm diam.	Fibres
Silver	10.5	1.59×10^{-6}	~100 μm	Spherical
			~100 μm	Spherical
Zinc	7.14	5.9×10^{-6}	~45 μm	Flake

Commercial polymer composites containing conductive carbon black make up greater portion of all the conductive resins available. Economics plays an important role since this conductive filler is the most cost effectiveness (Sichel, 1982; Novak et al.,

2002; Hu et al., 2003; Wan et al., 2005). In addition, by varying the concentration of the carbon black, conductive polymer composites that fall into the three different groupings, antistatic, static dissipative and conductive are made available (Das et al., 2002). Virtually all polymers can be compounded with carbon black.

As a general rule, physical strength properties of the base polymer are reduced when carbon blacks is added. The degree of loss is proportional to the concentration, the higher the loading, the lower the properties. However, in term of electrical properties, the result is reciprocal, where the increased in conductivity is observed.

2.2.4. Metal-coated filler

Metal-coated glass fillers have the advantage of the low density glass combined with the high electrical conductivity of the metal coating. Metal-coated glass fibres were investigated as a reinforcement for tires during 1950s. Metal coated glass fibres had been used in sheet moulding compounds and injections-moulded to produce an electrically conductive polymer composites having essentially the same mechanically properties as the unfilled polymers. The apparent key to achieving good adhesion between the metal coating and the glass fibres was the prior removal of the glass sizing. Glass fibre greatly increases tensile strength, flexural strength, and flexural modulus while at the same time reducing shrinkage.

Another approach to make conductive polymer composites is metal-coated carbon fibre or in particular known as nickel-coated fibre. This conductive polymer composite with very high conductivity, less than 1 ohm/cm volume resistivity, can be produced. The nickel-coated fibres are said to be 50 times more conductive than the carbon fibre. These products are excellent candidates for EMI/RFI shielding applications.

2.3. Type of Polymer Matrices

2.3.1. Thermosetting Resins

The most commonly used resins are epoxy, unsaturated polyester and vinyl ester (Kanchanomaia et al., 2005; Seyhan et al., 2007). These cover a very broad class of chemicals and a wide range of physical and mechanical properties can be obtained. In thermosetting polymers, the liquid resin is converted into a hard rigid solid by chemical cross-linking, which leads to the formation of a tightly bound three-dimensional network (Goodman et al., 1998; Schonbacher, 1998; Callister, 2006). This is usually done while the composite is being formed. The mechanical properties depend on the molecular units making up the network and on the length and density of the crosslink. The former is determined by the initial chemicals used and the latter by control of the cross-linking processes in the cure. Curing can be achieved at room temperature, but it is usual to use a cure schedule which involves heating at one or more temperatures for predetermined times to achieve optimum cross linking and hence optimum properties. A relatively high-temperature final post-cure treatment is often given to minimise any further cure and change in properties during service. Shrinkage during cure and thermal contraction on cooling after cure can lead to residual stresses in the composite.

2.3.2. Thermoplastics

Unlike thermosetting resins, thermoplastics are not cross-linked (Schonbacher, 1998; Callister, 2006). They derive their strength and stiffness from the inherent properties of the monomer units and the very high molecular weight. This ensures that in amorphous thermoplastics there is a high concentration of molecular entanglements, which act like cross-links and that in semicrystalline materials there is a high degree of molecular order and alignment. Heating of amorphous materials leads to disentanglement and a change from a rigid solid to a viscous liquid. In crystalline

materials heating results in melting of the crystalline phase to give an amorphous viscous liquid. Both amorphous and semi-crystalline polymers may have anisotropic properties, depending on the conditions during solidification. In amorphous polymers this is due to molecular alignment which occurs during melt flow in moulding or subsequently during plastic deformation. Similarly, in crystalline polymers, the crystalline lamellar units can develop a preferred orientation due, for example, to non-uniform nucleation at the surfaces of fibres, or in the flowing melt, and preferential growth in some directions because of temperature gradient in the melt.

Although thermoplastics cannot, in general, be expected to duplicate fully the mechanical behaviour of traditional metal alloys, a major effort has been undertaken in recent decades to produce some polymers with strength and stiffness to be serious candidates for structural application once in metals. These are known as engineering polymers, which retain good strength and stiffness up to 150-175 °C (Shackelford, 2000). The general use textile fibre nylon is also a pioneering example of an engineering polymer, and it continues to be the most important. It has been estimated that industry has developed more than half a million engineering polymer part designs specifying nylon.

2.3.3. Rubber

The term rubber originally meant the material obtained from the rubber tree *Hevea brasiliensis*. Today, a distinction is made between crude rubber and vulcanized rubber or elastomers.

Most of the dry natural rubber (NR) used today is obtained from the latex, a milky liquid that exudes from the bark of the tree when it is wounded or cut. The latter is coagulated and the solid materials separated, washed and dried to obtain a solid natural crude rubber (Nagdi, 1993). Later, man-made synthetic crude rubbers were

developed and became available in commercial quantities (Claude White, 2007). Synthetic crude rubbers are prepared by reacting certain low-molecular-weight substances called monomers to form long-chain molecules called polymers. They are usually obtained as a water emulsion known as synthetic latex, which is similarly coagulated and the solid material separated, washed and dried to obtain solid synthetic rubbers.

Crude rubber, whether natural or synthetic, are plastic-like materials that can be deformed at high temperatures. The elastic properties have to be developed by further compounding. The compounding means certain additives are incorporated in the crude rubber. When the rubber mixture is heated, a chemical reaction called vulcanization or cure takes place. In this process the chain molecules are fastened together at various points along their lengths by cross-links. These cross-links prevent slippage of chains pasty each other.

Unlike crude rubber, elastomers or vulcanized rubbers are elastic materials, that is, they have the ability to deform substantially under the application of force and then snap back to almost their original shape when the force is removed.

2.3.3.1. Natural rubber

Natural rubber (NR) is hydrocarbon consisting solely of carbon and hydrogen with an empirical formula of C_5H_8 with high molecular weight polymer of isoprene (Stern, 1982). It has a density of 0.93 units at $20^{\circ}C$ (Barlow, 1993). The chemical name for natural rubber is polyisoprene, which is a homopolymer of isoprene. It has the *cis*-1, 4 configurations as shown in Figure 2.1. In this polymer structure, two carbon atoms are joined together by a double bond and they are locked in a rigid structure. Due to the presence of an asymmetric carbon atom (two unlike groups attached to each of the carbon atoms), *cis* and *trans* forms are possible. If the two like groups attached to each

of the carbon atoms are in the same side, it called the *cis* form and if the two like groups are on opposite sides, the polymer is said to have the *trans* form. Natural rubber is a straight chain polymer structure which essentially all (90-95%) the isoprene have the *cis*-1,4 configuration (Fulton and Thorpe, 1996).

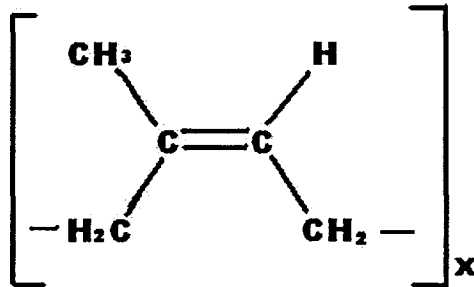


Figure 2.1: Polymer structure of *cis*-1,4 polyisoprene for natural rubber

SMR L grade natural rubber is introduced under Standard Malaysian Rubber (SMR) and the only rubber grade that has a colour specification. SMR L is natural rubbers graded by technical specifications, not by the conventional visual standards. These SMR L rubbers represent very high quality, clean latex grades of natural rubber.

2.3.3.2. Synthetic rubber

Synthetic rubber plants were built around the world after 1945, primarily in Europe, North America, and Japan. In 1960 use of synthetic surpassed that of natural for the first time.

Synthetic rubber is man-made and is produced around the world in manufacturing plants that synthesize it from petroleum and other minerals. Synthetic rubber is any type of artificially made polymer material which acts as an elastomer. An elastomer is a material with the mechanical (or material) property that it can undergo much more elastic deformation under stress than most materials and still return to its previous size without permanent deformation. Synthetic rubber serves as a substitute

for natural rubber in many cases, especially when improved material properties are needed.

Synthetic rubber can be made from the polymerization of a variety of monomers including isoprene (2-methyl-1,3-butadiene), 1,3-butadiene, chloroprene (2-chloro-1,3-butadiene), and isobutylene (methylpropene) with a small percentage of isoprene for cross-linking (Claude White, 2007). Furthermore, these and other monomers can be mixed in various desirable proportions to be copolymerized for a wide range of physical, mechanical, and chemical properties. The monomers can be produced pure and addition of impurities or additives can be controlled by design to give optimal properties. Polymerization of pure monomers can be better controlled to give a desired proportion of cis and trans double bonds.

An urgent need for synthetic rubber that is derived from widely distributed feed stocks grew out of the expanded use of motor vehicles, and particularly motor vehicle tires, starting in the 1890s. Political problems that resulted from great fluctuations in the cost of natural rubber led to enactment of the Stevenson Act in 1921. This act essentially created a cartel which supported rubber prices by regulating production. By 1925 the price of natural rubber had increased to the point that companies such as DuPont were exploring methods of producing synthetic rubber to compete with natural rubber (Synthetic rubber, 2007). In the case of Dupont the effort lead to the discovery of Neoprene which is a synthetic rubber that is too expensive to be used in tires, but has some very desirable properties that make it possible to use rubber in applications that would be unsuitable for natural rubber.

EPDM rubber (ethylene propylene diene monomer rubber) is one of synthetic rubber which is characterized by wide range of applications (EPDM rubber, 2007). Figure 2.2 shows the structural formula of EPDM with ENB as a diene. EPDM is a

synthetic rubber whose principal components consist of the compounds ethylene and propylene. A flexible rubber matrix forms when a small amount of diene is added to the mix. EPDM is available reinforced or unreinforced with both commonly used; it's also available in either a cured (vulcanized) or uncured (non-vulcanized) state. Vulcanized EPDM is the most common with non-vulcanized often used for flashing purposes. EPDM rubber is used in automotive weather-stripping and seals, glass-run channel, radiator, garden and appliance hose, tubing, belts, electrical insulation, roofing membrane, rubber mechanical goods and plastic impact modification.

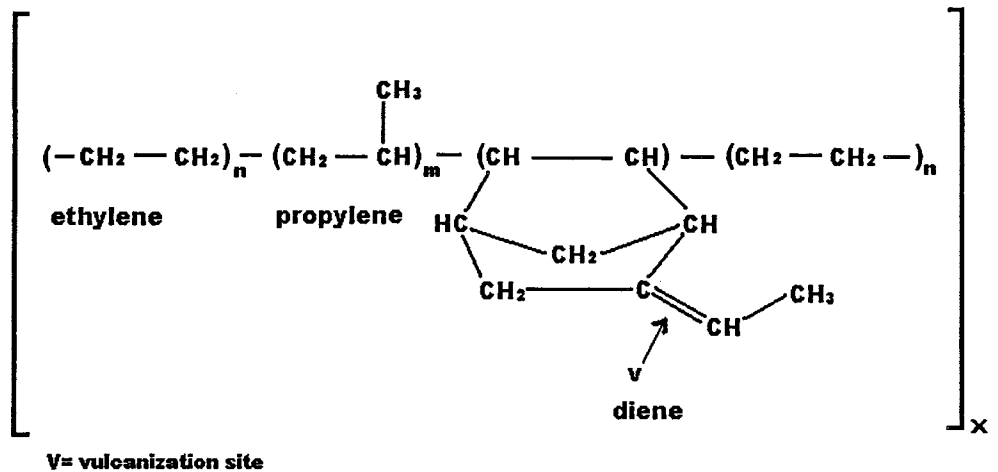


Figure 2.2: Structural formula of an EPDM with ENB

As non-polar elastomers, EPDM have good electrical resistivity, as well as resistance to polar solvents, such as water, acids, alkalies, phosphate esters and many ketones and alcohols (Nagdi, 1993). Amorphous or low crystalline grades have excellent low temperature flexibility with glass transition points of about minus 60 °C. Compression set resistance is good, particularly at high temperatures, if sulfur donor or peroxide cure systems are used. These rubbers respond well to high filler and plasticiser loading, providing economical compounds. They can develop high tensile and tear properties, excellent abrasion resistance, as well as improved oil swell resistance and flame retardance.

2.3.3.3. Degradation of rubber

In routine technological evaluation, rubber vulcanizates are subjected to accelerated ageing tests to get information about the service life. Natural rubber (NR), unlike many other polymers, is highly susceptible to degradation, due to the presence of double bonds in the main chain. Degradation of NR is accelerated mainly by heat, humidity, light, ozone, radiation etc. Many of the chemically unsaturated rubbers are prone to attack by even the minute quantities of ozone present in the atmosphere. Degradation of NR by radiation is also a serious problem, mainly from gamma ray irradiation, which is usually used in sterilization. With gamma ray irradiation high molecular weight materials may decompose (Vinod et al., 2002).

Ethylene propylene diene monomer rubber (EPDM) are valuable for their excellent resistance to heat, oxidation, ozone and weather aging due to their stable, saturated polymer backbone structure. Heat aging resistance of EPDM up to 130°C can be obtained with properly selected sulfur acceleration systems and heat resistance at 160°C can be obtained with peroxide cured compounds (Nagdi, 1993).

2.4. Type of conductive polymer composites

The ability of polymers when used as matrix can enhance the adhesive properties, corrosion resistance, processing characteristics, and strength to weight ratio of composites while maintaining excellent electrical and thermal insulations. There are certain applications, in which would be desired polymers which possess high electrical and thermal conductivities. The electrical and thermal properties of polymers can be increased by adding conductive fillers such as metals, carbon blacks, graphite etc (Zhao et al., 2007). By adding conductive filler to polymers, composites can be designed with specific properties tailored to each application. In order for composites materials to be used for conductive applications, the composites should have an

electrical resistivity in the range of 10^{12} and $10^8 \Omega \text{ cm}$ for ESD applications, 10^8 and $10^2 \Omega \text{ cm}$ for moderately conductive applications and 10^2 and lower for shielding applications (Boudenne et al., 2005). There are numerous examples where conductive filler have been added to plastics to produce conductive composites. Metal fibre/particles, including aluminium, steel, iron, copper and nickel-coated glass fibres have been used. Carbon black and carbon fibre have also been used to improve electrical properties.

The conductive fillers, such as carbon fibres and metal fillers, act as channels for the electron to flow through. The electrons are free to flow through the fillers and fibres. For example, in case of fiber, once the electron reaches the end of fibres, they encounter the polymers matrix, which act as dam, blocking the flow of the electrons. Once enough filler has been added, the carbon fibres begin to come in contact with each other, forming a complete path for the electrons to travel throughout the full volume of the composites.

Conductive carbon black polymer composites have been used for ever one-half century. Basically, carbon black was made from channel process, but today nearly all blacks are formed from oil by furnace process. When classified according to electrical resistivities, conductivities of polymer composites may differ, depending on the type of polymers and carbon blacks, its fraction of carbon loadings, particle size, the aggregate structure, and its inherent porosity. Conductive carbonaceous fillers of current commercial relevance include carbon black (CB), graphite (G) and carbon fibre (CF), which differ greatly in both structure and form.

Thongruang et. al (2002) have studied about correlated electrical conductivity and mechanical property analysis of high-density polyethylene filled with graphite and carbon fiber. They found that the addition of carbon fiber to HDPE/G composites

increase the conductivity relative to that of HDPE/G composites at the same filler concentration. The increase in conductivity is depended on carbon fiber length and becomes more pronounced at and beyond the threshold loading of the HDPE/G composite. Krupa et al. (2004) also used graphite filled polyethylene in their studied. They have focused on electrical and thermal conductivity and mechanical properties of the conductive composites. In their study, they have compared two types of polyethylene (LDPE and HDPE) as a matrix. They found that the electrical properties for LDPE and HDPE increased with increasing graphite loading. However, the degree of crystallinity of polyethylene matrix did not have a significant influence on electrical conductivity of composites. The thermal conductivity of filled HDPE is higher then thermal conductivity of filled LDPE in the whole concentration range due to higher degree of crystallinity of high density polyethylene.

A number of metals may be added to polymers to give conductive composites materials. Some metals have been added to enhance the electrical and thermal conductivity and also increase density, display permanent magnetism, and improve thermal stability. Nonconductive polymers exhibit volume resistivities in the range of 10^{11} to 10^{17} $\Omega\cdot\text{m}$. Conductive polymer composites possess the capability to have tailored resistivity ranging from 10^{10} to as low as 10^{-3} $\Omega\cdot\text{m}$, depending on such variables as the conductive type, aspect ratio, concentration, plastic material, and processing method (Strong, 1996). Not all polymer composites containing high loadings of metal powders are conductive, for example 200 phr of aluminium in epoxy resin may only reduce the resistivity from 10^{14} to 5×10^{12} $\Omega\cdot\text{cm}$. On the other hand, the resistivity as low as 3×10^{-4} $\Omega\cdot\text{cm}$ could be attained with 77% by weight of silver and that silver powders much less efficient in promoting conductivity than silver flake.

Nurazreena et al. (2006) found that at least 30 and 40 % by volume of aluminium fillers are needed to induce conductivity in a high density polyethylene

(HDPE) matrix. They investigate a wide range of metal fillers including copper and iron. They found that there is no significant increase in the conductivity of HDPE-Cu and HDPE-Fe between 30 and 40 % volume percentage are observed. They also found that the flaky shape of aluminium provide more stable conducting composite than non-dimensional particles (copper and iron) because of their large surface area per unit volume, therefore it imparts better metal to metal contact.

Electrical and thermal conductivity, diffusivity, effusivity and specific heat of polypropylene matrix filled with copper particles of two different sizes have been investigated by Boudenne et al (2005). The highest heat transport ability was observed for the composites filled with the smaller particles. Electrical conductivity investigations showed that the size of fillers also influences the percolation threshold.

2.5. Properties of conductive polymer composites

2.5.1. Electrical properties

The primary reason for adding conductive fillers to a polymer is to enhance the electrical and thermal properties of the polymer. Normally high insulating materials, polymers filled with conductive fillers can become excellent conductors of electricity. The change of a polymer from insulator properties to electrical properties is the most important to focus. The addition of conductor materials to insulator materials affects the electrical properties of the composite according to the degree of filling and proximity of conductive particles to other conductive particles. There is three situations are possible to occur after the addition of conductive materials to polymer; no contact between conductive particles, close proximity and physical contact.

When the conductive particles are isolated or no contact between conductive particles, the conductivity of the composite is changed only slightly. The composite

remains an insulator, although its dielectric properties may change significantly. When the conductive particles are in close proximity, electrons can jump the gap between the particles, creating current flow. The ability of an electron to jump a gap, under a given voltage field, increases exponentially with decreasing gap size. The size of the gap that electron can be jumped is about 10 nm.

The third condition which is possible when conductive filler is introduced into an insulating matrix is that the conductive particles will physically contact each other to form a continuous network throughout the composite. Under this condition the composite conducts through the particle network by the conduction mechanism of the particles. The behaviour of composites made with an insulating matrix and conductive fillers is quite interesting and extremely important to understand. The reason for these comments is that the composite changes from an insulator to a conductor over a very range of filler concentration. A typical curve of resistivity versus volume concentration of filler is shown in Figure 2.3. At low filler concentration the composite remains an effectively insulator. At a critical volume fraction, the composite's resistivity fall sharply to a level at which the composite can readily conduct electricity (Bhattacharya, 1986). Further increased in filler content above critical loading do not appreciably reduce the resistivity of the composite.

Conductive polymers can undergo a sharp transition from insulator to a conductor at a critical volume concentration of metal particles. Since the first report, many other researchers have shown the sharp transition from insulator to conductor. Different researchers have reported different critical volume concentration. Nurazreena et al. (2006) found that a composite of randomly dispersed aluminium fillers in a HDPE matrix became conductive at a volume concentration of 40 %. However, Tee et al. (2007) was reported that the transition regime from low to high conductivity or

insulator–conductor transition was found to occur at a range of critical volume fraction which is at 5 vol. % of silver filled epoxy composites.

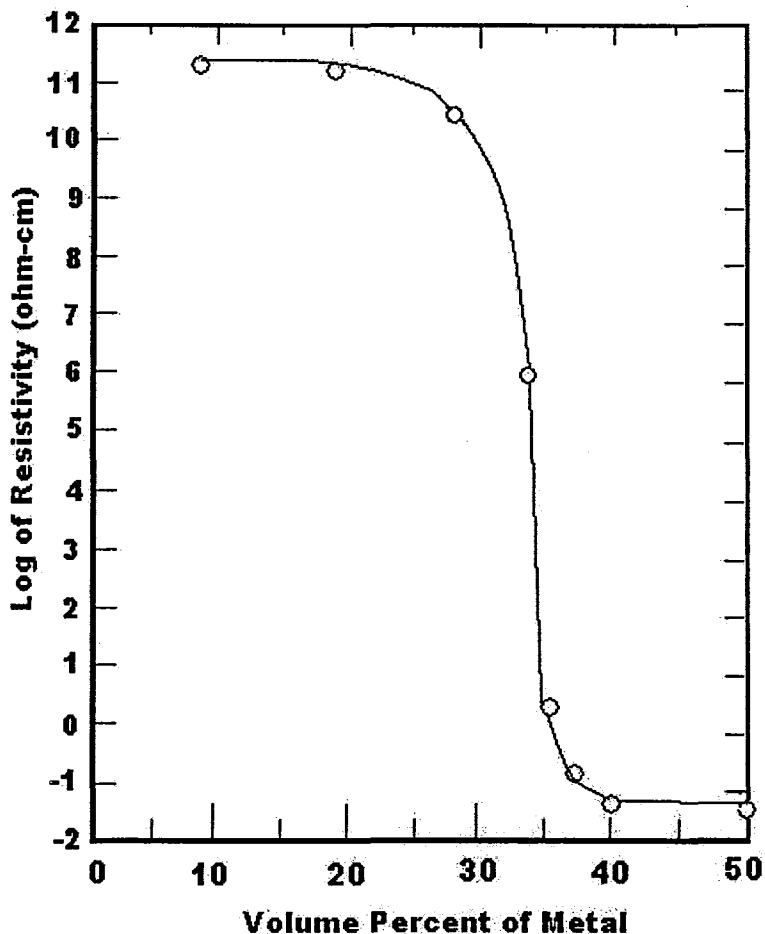


Figure 2.3: Resistivity versus volume fraction of silver spheres in Bakelite. (Bhattacharya, 1986)

Thongruang et al. (2002) have studied the variation in conductivity of HDPE composites containing graphite and the two carbon fiber lengths. They found that the composites containing the longest fibers exhibit the lowest threshold concentration at 7.5 wt% compared with 12.5 wt% for the shortest fibers and 50 wt% for graphite filled HDPE composites. In case of graphite, it consists of small primary particles that tend to aggregate into larger low-aspect-ratio cluster. It tends to disperse more evenly into polymer matrix, thereby resulting in fewer particle-particle contacts at a given concentration level and, consequently, a lower conductivity polymer composite.

2.5.2. Mechanical and thermal properties

The uses of reinforced polymeric materials are more desirable on mechanical and thermal properties. Intrinsic properties of polymers combined with the advantages of the fillers have led the creation of many new materials. These new materials have one or a combination of excellent mechanical, thermal, or electrical properties at lighter weights than their predecessors.

Work to date on mechanical and thermal properties of polymer containing dispersed conductive fillers clearly indicates that not only are the material properties of the individual phases and percentage of filler important, but the size, shape, orientation, and state of adhesion between the filler and matrix are critical. The differences between conductive fillers are the usually higher values of elastic modulus, thermal expansion coefficient, and thermal conductivity. These differences can affect the mechanical properties in two ways. First, the filler-to-matrix elastic modulus ratio E_f/E_m is approximately 20-100 for non-metallic fillers and 200-1000 for metallic fillers. Second, the adhesion between the matrix and filler particles is more critically influenced in the case of metal-filled polymers.

Vinod et al. (2002a) have studied about mechanical and thermal properties of aluminium filled natural rubber composites. They found that the mechanical properties are increased after the addition of aluminium in natural rubber composites. They also reported that the thermal properties of aluminium filled natural rubber composites also increase after the addition of aluminium particles. Aluminium filled natural rubber composites also showed better thermal properties compared to other fillers like high abrasion furnace black (HAF), general purpose furnace black (GPF), silica and acetylene black.

Nurazreena et al. (2006) found that the tensile properties of metal filled HDPE composites is influenced by the shape of the filler, degree of crystallinity and the adhesion between metal fillers and polymer. They have reported that the tensile strength is decreased with increasing filler concentration but the Young's modulus is increased with increasing amount of filler loadings. The decreased in tensile strength was due to the filler agglomeration which reduce the adhesion between metal to polymer and increase the metal to metal contacts.

2.6. Factor Influencing Electrical Resistivity of Conductive Polymer Composites

Several factors that influence the electrical properties of conductive polymers composites include the type, concentration, aspect ratio, and conductivity of the additive, as well as polymers materials selected. In most cases, these factors are all interrelated and therefore must be addressed collectively to yield the most cost effective material selection (Stauffer, 1985).

a. Conductive Filler concentration

Basically, an increase in conductive fillers concentration in polymer decreases the resistivity (increase the conductivity) of the composites materials. Generally, resistivity does not decrease with increasing conductive fillers concentration until the critical threshold level is reached. At this level, the resistivity of the polymer composites decreases sharply with every small increase in conductive filler concentration. The further increase in conductive filler concentration, which pass the critical threshold level continue to lower resistivity but at a substantially lower rate. Therefore, the conductive filler concentration influences the resulting resistivity values. The actual concentration of a particular conductive filler requires in a specific applications is directly related to all

of the factors previously mentioned, that is, filler type, aspect ratio, material, and processing method (McQueen et al., 2004).

b. Conductive filler size

The conductive filler size can affect the degree of connection and distribution between conductive fillers in the composites. Theoretically, continuity of conductive fillers will decrease the electrical resistivity (increase electrical conductivity) for the polymer composites materials. The differences in conductive filler size distributions significantly affect the critical concentration at which the polymer composites become conductive (Bhattacharya, 1986). It is known that a broad conductive filler size distribution have a lower probability of contacting neighboring filler than those with narrow size distributions.

c. Filler electrical conductivity

Filler electrical conductivity is a major factor in determining the resultant electrical properties of conductive polymers. It also controls the critical concentration level. All other factors being equal, the higher the conductivity of the filler, the lower the concentration required to achieve a particular level of conductivity.

d. Conductive filler shape

The electrical resistivity of polymer composites is strongly dependent on the shape of the conductive filler. The most common form of conductive filler was either spherical or irregularly shaped isotropic particles. In the past few year, anisotropic conductive filler shapes such as in the form of fibres, flakes, and ribbons have become available.