EFFECT OF CARBON BLACK LOADING ON THE PROPERTIES OF GRAPHITE-EPOXY COMPOSITE FOR BIPOLAR PLATE

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This report is presented to fulfill part of the requirement for my Degree of Bachelor in Mechanical Engineering (Hons)

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DECLARATION

"I here declare that the result in this report is my own research except as cited in the references. The thesis has not been accepted for ant degree and is not concurrently submitted in candidature of any degree."

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APPROVAL

"I hereby declare that I have read this thesis in my opinion this report is sufficient in term of scope and quality for the award of Bachelor of Mechanical Engineering (with Hons.)"

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DEDICATION

I would like to dedicate my thesis to my beloved parents, Sahrani Bin Abas and Siti Sariah Binti Abd Rashid also my supervisor Prof. Madya Dr. Zulkefli Bin Selam

ABSTRACT

The performance of Polymer Exchange Membrane Fuel Cell (PEMFC) is depends to the bipolar plate. This research study the alternatives materials for bipolar plate which is graphite-epoxy composites. Different ratio of graphite (G) and carbon black (CB) polymer exchange membrane fuel cell has been mixed with epoxy as the binder to fabricate the conductive composites bipolar plate. The compositions of fillers and binder is 80:20 and the percentage of CB has been varying 5% to 30% from total percentage of filler. The material will be pre-mixing in the ball mill machine. The conductive composites will be fabricated by using hot compressing molding. To identify the properties of the composites bipolar plate, the electrical conductivity, flexural strength, hardness and density will be measured. The result from the tests will be analyzed to identify whether the properties of conductive composites were achieved the DOE requirement. Based on this experiment, the critical loading of CB content in the composites is 20% and the present of carbon black in the composites improved the properties of graphite-epoxy composites such as electrical conductivity, flexural strength, hardness and density.

ABSTRAK

Prestasi polimer pertukaran membran sel bahan api bergantung kepada plat dwikutub. Penyelidikan ini bahan alternatif untuk plat dwikutub iaitu komposit grafit-epoksi. Nisbah yang berbeza untuk gafit dan juga kabon hitam dicampur dengan epoksi sebagai pengikat untuk meghasilkan komposit plat dwikutub yang konduktif. Komposisi pengisi dan pemgikat ialah 80:20 dan peratus karbon hitam diubah 5% kepada 30% daripada peratus keseluruhan pengisi. Bahan tersebut akan di menjalani proses pra-campuran menggunakan mesin ball mill. Komposit konduktif tersebut akan dihasilkan meggunakan mesin tekanan besuhu tinggi. Bagi mengenalpasti ciri-ciri plat dwikutub komposit, ujian pengaliran elektrik, kekuatan lentur, kekerasan dan ketumpatan akan diukur. Hasil kajian daripada ujian tersebut akan dianalisis untuk mengenalpasti sama ada ciri-ciri komposit konduktif tersebut mencapai kehendak DOE. Berdasarkan eksperimen, kandungan maksimum karbon hitam ialah 20% dan kehadiran karbon hitam dalam komposit meningkatkan komposit grafit-epoksi seperti pengaliran elektrik, kekuatan lenturan, kekerasan dan ketumpatan.

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CNTs	Carbon Nanotubes
CB	Carbon Black
G	Graphite
EP	Epoxy
PEMFCs	Polymer Electrolyte Membrane Fuel Cell
DC	Direct Current
DOE	Department of Environment
CPC	Conductive Polymer Composite

CHAPTER 1

INTRODUCTION

1.1 Background

Proton-exchange membrane fuel cells or another name for polymer electrolyte membrane (PEM) fuel cells (PEMFC) is a very essential technology in extinguish pollution and minimize economic dependence on producing oil countries by burning fossil fuels convert to energy. It is also a type of fuel cell being produced mostly for transport applications because it has high efficiency to change chemical energy to electrical energy without harm the environment. The proton exchanges membrane fuel cells (PEMFCs) convert hydrogen and oxygen into electricity at low operation temperature under 100^{o}C. One of the important components of a fuel cell is a bipolar plate. It provides 70% to 80% of stack weight and up to 45% of the cost. Thus, the bipolar plate is the most important part to be studied on the cost or the material performance.

Bipolar plates (BPPs) are playing a very important role in which should fulfill many functions which is homogeneously distribute gases over the area of the cell, separate the fuel and oxidant gases and prevent leakage of gases, collect the current produced in the Electrochemical reactions, discharge the water produced, and ensure the mechanical strength of the stack. The materials to make the bipolar have a standpoint. The based composition of the composites is multi-filler material and binder material. In this study, the main filler will be Graphite (G) and Carbon Black (CB) is the second filler while Epoxy will be the binder. The BPPs must be considerable achieve a good performance for fuel cell application such as electrical conductivity, thermal conductivity, gas permeability, mechanical strength, corrosion resistance, and low weight. Department of Energy, DOE has specified the properties of a bipolar plate. The properties have been developed with information from automotive and energy companies and specifically the Fuel Cell Technical Team. The properties requirement shown in Table 1.1 should be satisfied for fabrication of a bipolar plate.

Table 1.1: Properties requirement for bipolar plate (Sources: The office of Energy Efficient and Renewable Energy (2010))

Property	Value
Electrical conductivity	> 100 [Scm^{-1}]
Thermal conductivity	> 10 [$W(mK)^{1}$]
Flexural strength	> 25 [MPa]
Shore hardness	> 50
Bulk density	< 5 [g/cm^{3}]

Graphite is the most commonly used material for a bipolar plate. Graphite has a good conductivity and excellent corrosion resistance with a low density of about 2 g cm^-3. The weakness of Graphite are lacks of mechanical strength and poor ductility. The metal bipolar plate is most frequently made of special alloyed steel due to its high strength, high chemical stability, ease of mass production and low-cost. However, stainless-steel are low surface contact resistance and inexpensive mass production. In this case, composites materials are the best solution. Various graphite-polymer composites and those on the basis of the inorganic binding agent are widely used.

Carbon black is materials that produce incomplete combustion of heavy petroleum products such as FCC tar, coal tar, ethylene cracking tar, with an addition of a small amount of vegetable oil. It is usually used for reinforcement of polymer, black pigment, and electrical conductivity. The main properties of carbon black are in Table 1.2. Carbon black present a complex morphology that could fill the holes left.

Table 1.2: Principal properties of Carbon Black (Sources: Planes, Flandin and Alberola, (2012))

	Carbon Black
Density (g.cm^{-3})	1.7 – 1.9
Particle Size	14 – 250 nm
Specific surface area (m^2/g)	7 - 560
Aspect Ratio	Close to 1
Conductivity ($S.cm^{-1}$)	2.5 - 20

1.2 Problem Statement

Polymer Electrolyte Membrane Fuel Cells (PEMFCs) is not a new thing to be studied. Many research has been done on the PEMFCs and most of the research is done on a bipolar plate by changing its pure graphite and based metal. There are few types of materials usually used to produce the bipolar plate. However, the weakness of pure graphite is brittle and low conductivity. The metal-based bipolar plate needs a good coating to prevent it from corrosion. This will increase the cost to manufacture the bipolar plate. The most common composites materials used to manufacture the bipolar plate is graphite because it has good electrical conductivity and excellent corrosion resistance with a low density of about 2 g cm^-3. The cons of graphite are low in mechanical strength and brittle.

Non-porous graphite is the references material in terms of durability and performance. This will make the bipolar plate brittle and allows gas permeation through the channels formed after machining. Post processing, such as resin impregnation need to be done because it is an efficient way to fill the holes, but the product cost will be increasing due to this process. Extensive efforts have been made to develop alternative materials, essentially metallic and composite bipolar plates.

An alternatives materials have been developed through composites approach where two type of materials combined, filler and binder. The materials used as filler are G, CB where graphite as the main filler and epoxy as the binder. These composites provide lower weight and the manufacturing process does not complex compared to traditional graphite.

1.3 Objective

The main objective of this research is:

1. To study the effect of Carbon Black (CB) on the properties of Graphite (G)-Epoxy (EP) composites.
2. Determine the critical loading of Carbon Black (CB) in G/CB/Epoxy Composites.

1.4 Scope of Project

This research will study the effect of CB loading on the electrical and mechanical properties of G/CB/EP composites. The critical loading of CB in G/Epoxy composites needs to be determined and to be used as the composition of G/CB/Epoxy composites for the bipolar plate. The materials will be used in this study are Graphite (G) as the first filler and Carbon Black (CB) as the second filler and Epoxy (EP) as the binder. Compression method will be the method of fabrication which saves cost and time. There are about five testing method, Electrical Conductivity test, Flexural Strength test, Bulk Density test, Shore Hardness test, and Microstructure test.

CHAPTER 2

LITERATURE REVIEW

2.1 Fuel Cell

A fuel cell is an electrochemical device that can convert the chemical potential energy of gaseous or liquid reactant into electrical energy as shown Figure 2.1. Fuel cells are widely used in power-driven handy equipment such as battery charges, laptops, external power units and electronic devices (Youssef, Amin and El-Khatib, 2018). The fuel cell has its own advantages which are rechargeable and environment-friendly. Fuel cell come in many varieties, however, they all work in the same general manner. There is two general type of fuel cell, low-temperature fuel cells, and high-temperature fuel cell. Low-temperature fuel cell are Direct Mechanical Fuel Cell (DMFC), Proton Exchange Membrane Fuel Cell (PEMFC) and Alkaline Fuel Cell (AFC). High-temperature fuel cells are Phosphoric Acid Fuel Cell (PAFC), Solid Oxide Fuel Cell (SOFC) and Molten Carbonate Fuel Cell (MCFC). Table 2.1 shows the characteristics of different type of fuel cells. This project will go through deeply about PEMFC and its main component, bipolar plate.

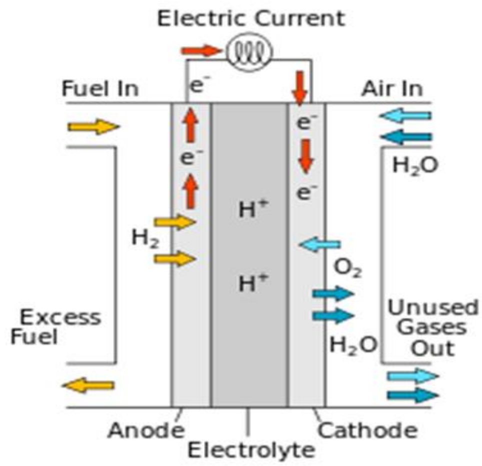


Figure 2.1: Basic fuel cell schematic diagram (Source: Wikipedia)

Table 2.1: Characteristics of different type of fuel cells (Source: Ismail, (2011))

		Electrolyte	Temperature Range	Fuel	Power Range	Application Example
Low-Temperature Fuel Cell	Direct Methanol Fuel Cell (DMFC)	Proton-conducting membrane	< 100 °C	Methanol	Watts/ kilowatts	Vehicle/ small appliances
	Proton Exchange Membrane Fuel Cell (PEMFC)	Proton-conducting membrane	< 100 °C	Hydrogen	Watts/ kilowatts	Vehicles/ small generator, domestic supply, power station

	Alkaline Fuel Cell (AFC)	Caustic Potash solution	< 100 °C	Hydrogen	Watts/ kilowatts	Outer space
High Temperature Fuel Cell	Phosphoric Acid Fuel Cell (PAFC)	Concentration phosphoric acid	~200 °C	Hydrogen	Kilowatts	Block type heat, power station
	Molten Carbon Fuel Cell (MCFC)	Molten Carbonate	~650 °C	Natural gas, coal	Kilowatts/ megawatts	Power plant combined heat and power
	Solid Oxide Fuel Cell (SOFC)	Ceramic	800-1000 °C	Natural gas, coal	Kilowatts/ megawatts	Power plant combined heat and power

2.2 Polymer Electrolyte Membrane Fuel Cells (PEMFCs)

The increased use of fossil fuel continues to draw an ever-increasing attention towards the development of alternative electrochemical energy technologies. In this regard, fuel cells are being considered as one of the most promising green and effectual energy conversion devices and PEMFCs is the best types of fuel cell because of the wide variety of power application ranging from portable and mobile applications to stationary power systems. The proton exchanges membrane fuel cells (PEMFCs) changes hydrogen and oxygen into electricity at low operation temperature. According to Youssef (2015), PEMFC achieves most of the portable fuel cell stack requirements application, because of its high energy density, long operation time, immediate refilling and the self-discharge. The Department of Energy (DOE) is focusing on the PEMFCs as the most important of vehicle applications. It able to efficiently to generate high power density and low operating temperature of about 60 to 80 degree Celsius. As shown in Figure 2.1, PEMFCs has anode and cathode which occur a chemical reaction to produce electricity. The chemical reaction at the anode and cathode can be explained through the chemical reaction as shown below:

Anode: $2H_2 \longrightarrow 4H^+ + 4e^-$

Cathode: $O_2 + 4H^+ + 4e^- \longrightarrow 2H_2O$

Overall: $2H2 + O2 \longrightarrow 2H2O$

2.2.1 Main Component of PEMFC

PEMFCs consist of many components and there is only four main component. There are Membrane Electrolyte Assembly (MEA), Bipolar Plate (BPs), End Plate and Collector (Maiyalagan and Pasupathi, 2010). An exploded view of PEMFC main component is provides in Figure 2.2. Hydrogen gas is fed to the fuel cell anode, which flows to the anodic catalytic layer where it is oxidized through the gas diffusion layer (GDL), resulting in proton transfer through the proton exchange membrane (PEM) and electron transfer through an external electrical circuit. Oxygen gas is fed to the cathode and upon diffusion to the cathodic catalytic layer, it combines with the protons and is reduced to water.

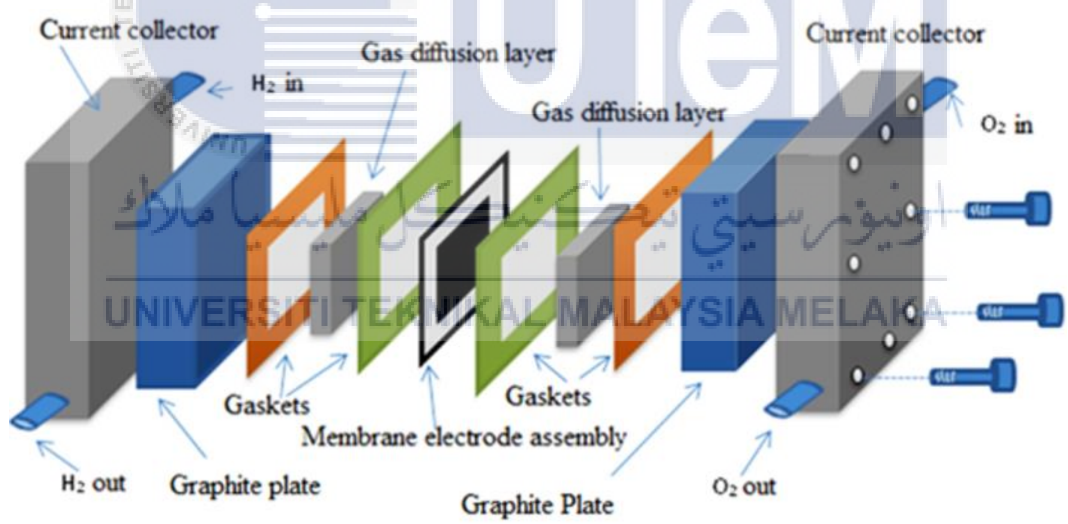


Figure 2.2: Exploded view of PEMFC main component

(Source: Wikipedia)

2.3 Bipolar Plate

According to Dobrovol'skii et al. (2007), an important component of a solid state FC is a bipolar plate (BPs) and end plate. Bipolar plates link together separate cells in a FC stack to form membrane electrode assemblies (MEAs) as shown in Figure 2.3. MEAs will control heat, gas, and electric flows. The bipolar plate contributes the majority of the weight and cost of PEMFC which about 80% of the total weight and 30% to 40% of the cost. Therefore the weight of bipolar plate needs to be light, smaller volume and low cost. Bipolar plate work in temperatures of about 100 °C and it can be higher. Therefore, the important characteristic of BPs are as below:

- specific electrical conductivity and heat conductivity
- corrosion and heat resistance at operating temperature
- stability in moisture rich condition
- chemical stability in the presence of reagents,
- lack of component poisonous for MEAs and low gas permeability

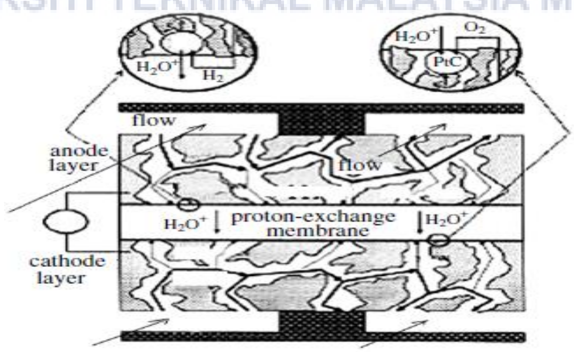


Figure 2.3: Structure of a membrane electrode assembly (MEA). The arrangement of principal units between gas flow along BP channels and flows in MEA

(Source: Yu. A. Dobrovol'skii and A. E. Ukshe)

2.4 Materials for Bipolar Plate

In creating a bipolar plate its need 3 common materials which are pure graphite, metal based and CPCs. A DOE technical target for PEMFC bipolar plate is provide in Table 2.2. Bipolar plate can achieve great mechanical and electrical properties if the amount of all materials controlled very well. Many research has been done on determining the best materials used to build bipolar plate. Graphite had been considered and extensively investigated for BPs application because it has excellent electrical conductivity and corrosion resistance. However, the machining process involved in producing graphite BPs take time and costly. Its brittle and porous nature structure makes the production of thin BPs to meet low cost and lightweight requirement impracticable. Therefore, research attention has shifted towards alternative material types that can counter the brittle and porous structure such as carbon-polymer composites (CPCs) and metals. (Alo *et al.*, 2017)

Table 2.2: DOE technical target for PEMFC bipolar plates (Sources: Alo et al, (2017))

Property	Unit	2017	2020
Cost	$\$KW^{-1}$	3	3
Weight	$KgKW^{-1}$	<0.4	4
H2 permeation rate	$cm^3(cm^3s)^{-4}$	$<1.3 \times 10^{-14}$	1.3×10^{-14}
Corrosion at anode, cathode	μAcm^{-2}	<1	<1
Electrical conductivity	Scm^{-1}	>100	>100
Area specific resistance	Ω-cm^2	0.02	0.01
Flexural strength	MPa	>25	>25

2.4.1 Pure Graphite

Pure graphite is a mineral form of the element carbon. It is extremely soft mineral and it breaks into minute, flexible flakes that easily slide over one another. Graphite is the only non-metal element that is a good conductor of electricity. Natural graphite is usually used in refractory applications which application that involve extremely high heat. According to Selamat et al., (2016), it stated that optimization for the highest electrical conductivity is increasing until 170^{o}C which is 50 S/cm^2 of electrical conductivity. Therefore, when the filler loading at 20 wt% the electrical conductivity will be in the highest state.

2.4.2 Metal Based

Metal based is categorized as a common and inexpensive metal. It may be discriminate by oxidizing and corroding relatively easily and reacting variably with diluted hydrochloric acid, HCl to form hydrogen.

2.4.3 Conductive Polymer Composite (CPCs)

CPCs is a combination of the polymer matrix which is thermoplastic or thermosetting and conductive filler, such as graphite (G), expanded graphite (EG), carbon black (CB), carbon fiber (CF), carbon nanotubes (CNTs) and graphene, to increase the electrical conductivity. The final properties of the composites depend on the polymer-filler combination. Thermoset-matrix composites usually have the advantages of higher strength and resistances than the thermoplastic-matrix composites (Alo et al., 2017)

2.5 Materials

The materials to be focus to produce the CPCs are G, CB powder as the filler and thermoset materials which is epoxy as the binder. Thermosets can be loaded with greater levels of filler during processing, compared to the thermoplastic. The example of thermosets that is commonly used in BPs is epoxy resins, phenolic resin, and vinylesters. The advantages of CPC bipolar plate are good corrosion resistance, lightweight, low cost, and ease of machining or molding gas channels during processing.

2.5.1 Filler

Polymer is commonly electrical insulator, so the main reason the additive of filler is to improve the electrical and mechanical properties of a bipolar plate. It is about two combination of filler which is G and CB and this will make the fuel cell have the good conductivity characteristic.

2.5.1.1 Carbon Black (CB)

Carbon black is composed of fine particles consisting mainly of carbon but it is different from graphite and carbon fibers because of aggregates having complex configurations, quasigraphitic in structure and of colloidal dimensions. Various features of carbon black are controlled in production by partially combusting oil or gases. Carbon black is widely used in various applications from black coloring pigment of newspaper inks to electric conductive agent of high-technology materials. It is used mainly in tires as excellent rubber reinforcement. Carbon black is also an excellent coloring agent as black pigment, and therefore is widely used for printing inks, resin coloring, paints, and toners. Furthermore, carbon black also used in various other applications as an electric conductive agent, including antistatic films, fibers, and floppy disks.

2.5.1.2 Graphite

According to Planes, (2012), graphite is the most crystalline form of carbon, apart from diamond and fullerenes. It has the properties both of metal such as thermal and electrical conductivity and of a non-metal such as inertness, high lubricity and thermal resistance. Table 2.3 shows the graphite properties.

Table 2.3: Principal properties of Graphite (Sources: Planes et al. (2012))

	Graphite
Density (g.cm^{-3})	2-2.25
Particle size (µm)	6-100
Specific surface area (m^2/g)	6.5-20
Aspect ratio	Close to 1
Conductivity (S.cm^{-1})	400-1250

The Graphite layers are bonded by weak van der Waals forces and an intercalation of atoms and molecules is possible between the graphene plans. Natural graphite with a layered structure can be converted to graphite oxide (GO) or graphite intercalation compound (GIC). This can be done through chemical oxidation than to expanded graphite (EG) with the action of temperature. Expanded Graphite expansion induces a modification of spacing between graphite layers, inducing a downsizing of the density between 10^{-3} and 10^{-2} g.cm^{-3}, whereas increasing the surface area with the mean ratio aspect to 40 m2.g^{-1} and 15 respectively. Moreover, the electrical properties are improved, since a conductivity of 12500 S.cm^{-1}. The "paper" materials based on GNPs have been reported with conductivities as high as 350 S.cm^{-1} much above the needs for the BPPs application.

Derieth et al., (2008) have stated that the electrical conductivity of a bipolar plate is influenced by the size and morphology of graphite particles. There are three set of graphite materials, the exact equal morphology but different sizes and spherical in morphology. The graphite with the same morphology is flake like and high aspect ratio while the graphite with spherical in morphology has low aspect ratio. All pure graphite materials were compounded under identical process conditions, with and equal filling load and the binder is polypropylene. It was found that the electrical conductivity of flake like particles increased as the particles size was reduced and also more conductive composites than spherical in morphology.

2.5.2 Binder

There are 2 types of binder, thermoset and thermoplastic. The benefits of using thermosets-matrix are during processing, their viscosities are low and they thus can be loaded with a high level of conductive filler and low density combined with good corrosion resistance. It also has dimensional and thermal stability. There are three types of thermosets mostly used in the fabrication of bipolar plate, epoxy, phenolic and vinylester resins.

2.5.2.1 Epoxy

The other name for epoxy is polyepoxides. Epoxy is a thermoset binder. Epoxy will be the materials that act as a glue for the composites. The maximum percentage of epoxy is 20% to achieve the maximum electrical conductivity.

2.6 Fabrication Process

There is some fabrications method that can be used in fabricating the bipolar plate for example stamping, die casting, investment casting and other. Most of the researcher has used injection molding and compressing molding and this makes that two method popular in developing the polymer composites bipolar plate. Both methods have their own advantages and disadvantages.

2.6.1 Injection Molding

Injection molding is one of the manufacturing processes for producing parts. It works by injecting molten material into a mold. All the materials of the part are fed into a heated barrel and mixed. Than being injected into a mold cavity, where it cools and hardens to the configuration of the cavity. This method is more convenient and interesting due to the high productivity inherent to the process that favors massive production rates but low production cost.

However, the process will be difficult because of the increasing viscosity of the mass due to the high loadings of conductive fillers. T. Derieth has stated that injection molding is the standard production method to manufacture bipolar plate in a one-step process directly with the required structures. The high content of the filler results in high viscosity and high flow resistance of the mass.

Mighri et al (2004) used injection molding as the method in manufacturing polymer composites bipolar plate. The materials are Polypropylene (PP) and Polyphenylene Sulphide (PPS) filled with graphite, carbon black and carbon fiber up to 60 wt%. The highest conductivity that can be reached by the bipolar plate is 17 Scm^{-1}.

2.6.2 Compression Molding

Compression molding is a well-known technique to develop a variety of composites products. It is a closed moldings process with high pressure applied during processing. In compression molder, the base plate is stationary while upper plate is movable and act as a puncher. Reinforcement and matrix are placed in the metallic mold and the whole assembly is kept in between the compression molder. The first step is prepared the powder compound in an extruder or an internal mixer at the proper temperature. This compound is then fed into a heated mold flows due to the application of pressure and heat and acquires the shape of the mold cavity with high dimensional accuracy which depends upon mold design.

Boyacı San and Okur, (2017) studied the effect of compression molding parameters on the electrical and physical properties of the polymer composites bipolar plate. The maximum electrical conductivity of 107.4 Scm^{-1} is obtained when the temperature reaches 187 ^{o}C with the pressure of 119 bar and time of 5 minutes. While the surface roughness is 0.74 µm at given conditions. The fabrication method has made the bipolar plate fulfilled the electrical conductivity target of DoE.

Dhakate et al., (2007) develop a graphite composite bipolar plate by using a compressing molding technique. The composites bipolar plates is added with some other reinforcing filler constituents, synthetic graphite of few micron sizes, carbon black of few micron particles sizes and carbon fibers of length less than 1mm with diameter 7 µm. Two types of phenolic resins are used as polymer matrix which is resoled type in liquid form and novolac in powder form. Pressure applied to the contact molding is 100 kg cm^{-2}. The carbon materials in its different structural form give different independent properties and it is proved that no single reinforcing filler constituent based composite plate gives the necessary properties for being used as bipolar plate in the PEM fuel cell.

2.7 Testing Method

There are several testing will be going through to know the properties of the bipolar plate. All the test is carried out to determine the electrical and mechanical properties. The testing is electrical conductivity testing, flexural properties testing and others.

2.7.1 Electrical Conductivity Testing

Graphite and metals have conductivity values which are enough for bipolar plate applications. However, a polymer composites bipolar plate will affect the conductivity because the polymer matrices generally have low conductivities. A bipolar plate that has higher filler contents can achieve high conductivity. The factors that affect the electrical conductivity of CPC bipolar plate are the type of matrix, filler type, particle size, orientation and dispersion, and processing technique. (Alo et al., 2017)

Electrical conductivity testing is a measure of the ability of the materials allowing the transport of an electrical charge or the ratio of the current density to the electrical field strength. Lee et al., (2009) have made the electrical conductivity test on the CPC bipolar plate. The electrical conductivity was measured using the four-point probe method according to the ASTM D4496 procedure. The programmable current source which is Keithley 6220 was used to apply the current (10mA) and to measure the voltage drop a Keithley 2182A nanovoltmeter was used. The averaged value among 10 readings for each sample was taken as the conductivity of the composite. The resistivity (ρ) and conductivity (σ) were calculated as follows:

$$\rho = \frac{\mathrm{RWT}}{L}, \qquad \sigma = \frac{1}{\rho} = \frac{L}{RA} = \frac{L}{RWT}$$

Where R is the electrical resistance. L, W and T are the length, width and thickness of the specimen and A is the area of the plane.

2.7.2 Mechanical Properties Testing

Mechanical tests that are usually carried out on the bipolar plate are a tensile test, compression test, impact test and flexural test. However, the test that will be focus on is flexural test. According to Alo et al (2017) a flexural test usually measured using the three points bending test. Metal generally sufficient the mechanical properties for bipolar plate application but for the CPC bipolar plate, a major challenge is the degradation in mechanical strength when the filler content is raised to levels needed to impart sufficient electrical conductivity. Some researchers have produced CPC bipolar plate with high electrical conductivity and also additional CFs to ensuring the strength.

CHAPTER 3

METHODOLOGY

3.1 Introduction

This chapter will be focus on how the conductive polymer composites bipolar plate is produce starting the fabrication method until the testing method. Experiment overview will be the first topic to be discuss and followed by raw materials preparation, pre-mixing of raw material, fabrication method, mixing process, compression molding and the last one is testing properties and analysis.

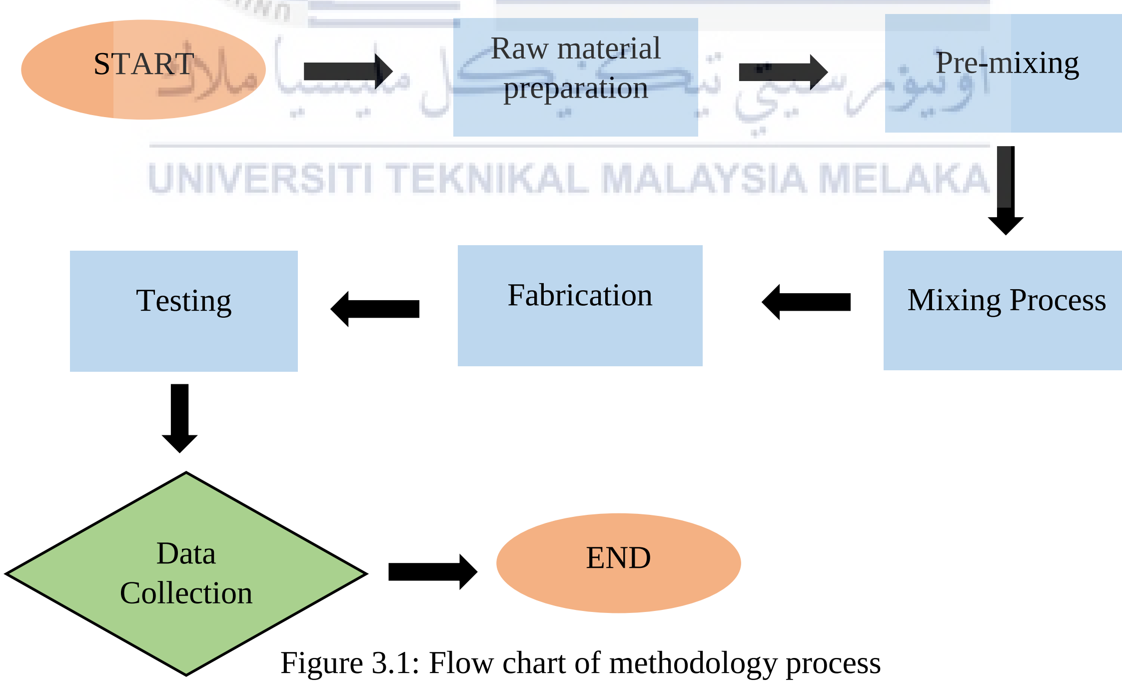


Figure 3.1: Flow chart of methodology process

3.2 Raw Materials

There are two type of materials will be selected in this research to fabricate the bipolar plate, Polymer matrix and Conductive fillers. G and CB will be the conductive filler, while EP will be the polymer matric which act as a binder for the CPC bipolar plate. The reason all the materials choose is due to its corrosion resistance, easy to get and low manufacturing cost. Table below show the characteristic each of the materials.

Table 3.1: Materials properties of G/CB/EP (Sources: Selamat et al. 2013)

Materials	Grade	Density (g/cm^3)	Thermal stability (^oC)	Size	Resistivity
G	3243	1.74	3500-4000	≤ 60 µm	0.036 Ωcm
CB	5303	1.7-1.9	3000	≤ 5 µm	0.314 Ωcm
EP	105/206	1.15	180-220	-	1014 Ωm

3.3 Pre-mixing

Pre-mixing process is mixing process of both filler materials, G and CB. Both materials is mix using ball mill machine shown in Figure 3.2. The mixing process is done about 1 ½ hour. The ball mill machine is a cylindrical device used to mix raw materials of graphite and carbon black by rotating around a horizontal axis partially filled with materials to be mix and also the mixing medium (steel ball). Ball mill capable to crush and transform large materials into fine powder.

Figure 3.2: Ball mill

3.4 Mixing Process

This process is the next step when the graphite and carbon black is mixed and form into powder. In this process, the mixed graphite and carbon black will be mix with epoxy by using high speed mixer for stirring purpose. The purpose of this mixing process is to make the composites of the materials of mixed graphite-carbon black with epoxy. The mixing process is about one to two hours and its stop until the compound mixture become homogen.

3.5 Fabrication

There are many method of fabrication the bipolar plate. Most of the researchers used injection molding and compression molding method in manufacturing CPC bipolar plate. In this research, compression molding will be method in fabricating the bipolar plate.

3.5.1 Compression Molding

This process is using Hot Press Machines. The mixed graphite-carbon black epoxy composites is pour into the mold carefully and immediately after internal mixing process is done

During the compressing process, the parameters need to be considered and listed are show in the Table 3.2.

Table 3.2: Parameters for hot pressing

Parameters	Value
Load	49 tones
Temperature	80 ^{o}C
Pre-heating time	6 minutes
Pressing time	30 minutes
Curing time	Pressure drop (80 ^{o}C – 50 ^{o}C)

The mold is a rectangular shape with a dimension 100mm x 100mm as shown in Figure 3.3 (b). When the compound material were put in the mold, the then put into the hot compression molding machine as shown in Figure 3.3 (a).

Figure 3.3 (a): Hot press machine

Figure 3.3 (b): 100mm x 100mm square mold

3.6 Testing Method

When the fabrication of the compound is completed, the compound must be analyzed to know the effect of carbon black on the properties. Therefore, several testing need to be carried out to know the properties of the bipolar plate. There are four testing, electrical conductivity, density and hardness testing, and flexural strength testing.

3.6.1 Electrical Conductivity

In this testing, the fabricated bipolar plate will be used Jandel Multi Four Point Probes technique at constant current supply to measure the resistivity of the sample. Figure 3.4 shows the Jandel Multiheight Microposition Probe use for electrical conductivity testing. If the electrical conductivity of the bipolar plate is higher than 100 Scm^{-1} that's mean the bipolar plate reached the DOE target. This test will be carried out based on F390-98 standard.

Figure 3.4: Jandel Multiheight Microposition Probe

3.6.2 Bulk Density

Density of the CPC bipolar plate is one of the important parameters need to be measured. To make sure the bipolar plate easy to transported and suitable for electronic device usage, the bipolar plate must be light. In this testing, Electronic Densimeter machine will be the device to test the density of the bipolar plate. According to DOE the strength of the bipolar plate must me more than 25 MPa and the weight must not exceed 0.4 kg/kW based on D1895 standard. Figure below show the Electronic Densimeter machine.

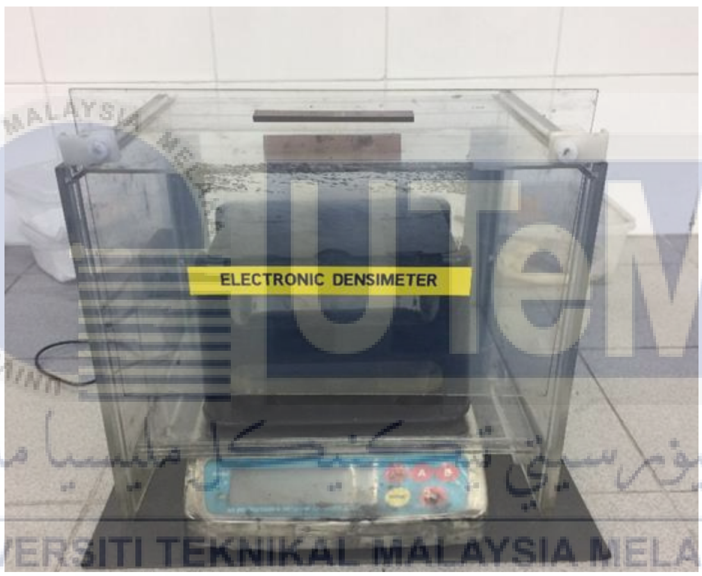


Figure 3.5: Electronic Densimeter

3.6.3 Shore Hardness

This testing will use stereoscopic tester to measure the shore hardness of the CPC bipolar plate. Its involved dropping a diamond tipped hammer, which fall inside a composites plate under the force of its own weight from a specific height onto the test specimen. This test conducted under D2240 standard.

Figure 3.6: Shore hardness tester

3.6.4 Flexural Strength

The flexural strength of the bipolar plate will be measure using three-point method. The machine to apply the method is Instron Universal Testing Machine. The maximum load will be set to 10 kN with the support span is 70 mm and the speed of machine is about 2 mm/ minutes. D790 standard will be used during the test.

CHAPTER 4

RESULT AND ANALYSIS

In determining the properties of the CPC bipolar plate, several tests on it needs to be done after the process of fabrication is fully complete. All the result from the testing process will be discussed and analyzed based on the different percentages of the Carbon Black in each six sample.

4.1 Electrical Conductivity

Jandel Multi Height Four Probe is the device to carry out the electrical conductivity testing in determining the ability of the CPC bipolar plate to conduct electricity. 18 reading was taken, 19 from the top surface and the other 19 from the bottom surface. The reading is calculated using the below formula to get conductivity in Siemens per Centimeter (S/cm).

$$\mathbf{S\ cm^{-1} = \frac{1}{2\pi S\ (data\ average)(0.6336)}}$$

Where: S = 0.1cm (distance of Jandel Multi Height Four Point Probe)

0.6336 = factor of thickness per diameter per specimen

Table 4.1 shows the electrical conductivity result of each specimen based on different percentages of CB. While figure 4.1 shows the plotted graph of electrical conductivity against the percentage of CB.

Table 4.1: Conductivity of G/CB/ EP bipolar plate

Carbon Black content (wt %)	**Conductivity (S/cm)**
5	4.66
10	12.43
15	17.05
20	21.84
25	19.96
30	19.83

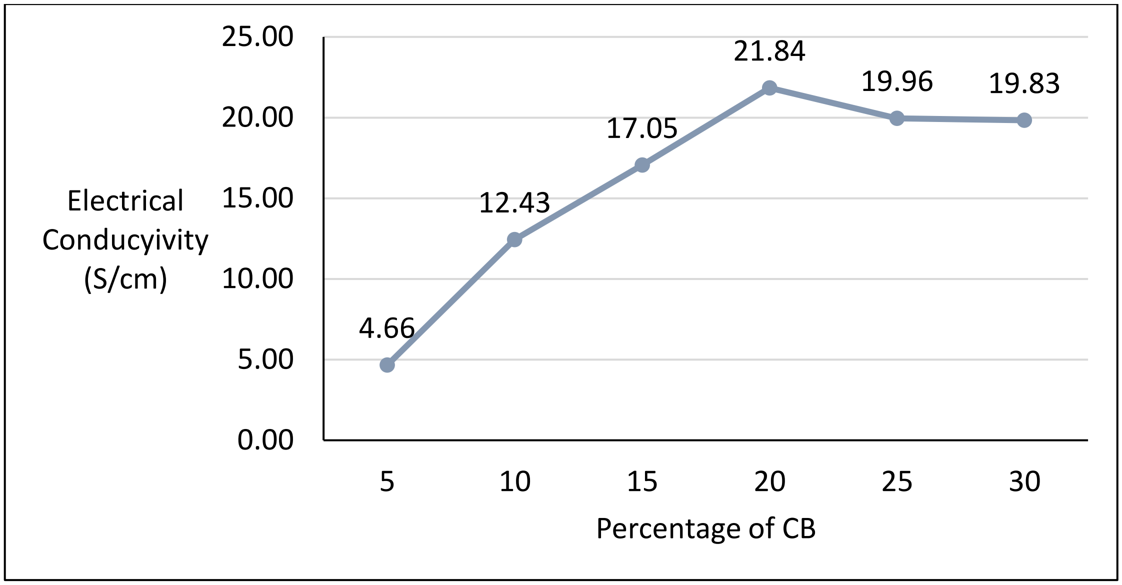


Figure 4.1 Graph of electrical conductivity against the percentage of CB

Based on the threshold percolation theory, the increasing of filler content will increase the electrical conductivity of the bipolar plate due to the continuous conductive network that is formed between the filler. However, there is a limit for the CB content in that filler. Based on the graph above, the critical loadings of CB in the composites is 20%. The bipolar plate specimen with 20% CB content has the highest value of Electrical conductivity. After 20% onwards, the electrical conductivity of the bipolar plate decrease because there is no longer conductive network are produced. The graph shows that GP/CB/EP bipolar plate meets the threshold percolation theory but all the specimen does not meet the standard conductivity value set by the US-DOE which is 100 S/cm or more.

4.2 Flexural Strength

Figure 4.2 shown the graph of flexural stress against the percentage of CB shows that the highest flexural stress is 10.097 MPa with the carbon content 10%. The flexural stress value starts to reduce when the percentage of CB content increase due to the lack of resin content that is work as the binder between filler and resin. This result does not meet the standard set by the US-DOE which is 25 MPa or exceed.

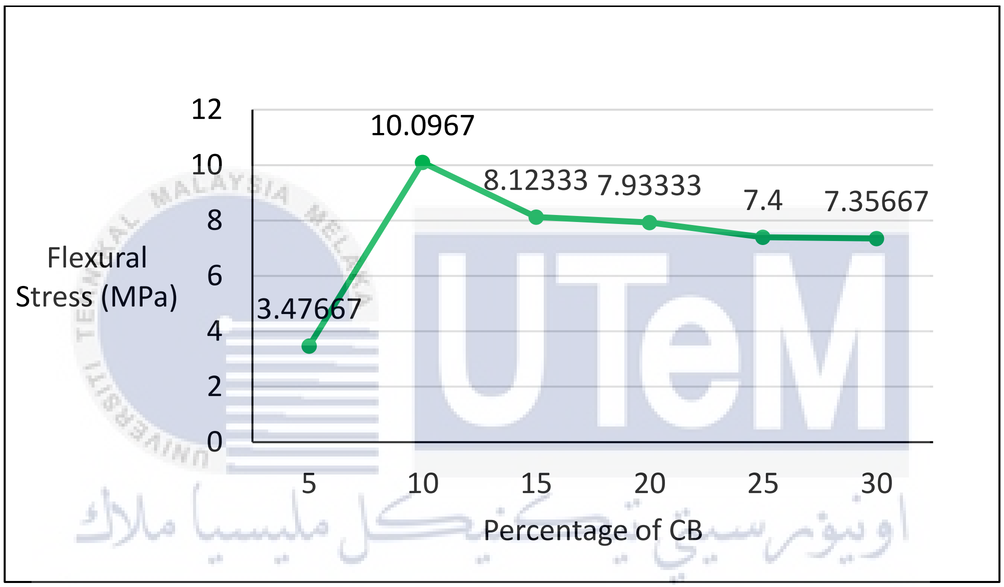


Figure 4.2 Graph of flexural stress (MPa) against the percentage of CB

4.3 Density

Figure 4.3 shown the graph of density against the percentage of CB above shows the specimen that has the highest density value is specimen with the CB content 0%, 1.73 g/cm^2. The density of the G/CB/EP bipolar plate decrease when the content of CB increases. All the specimen meets the US-DOE requirement for the bipolar plate which is less than 2 g/cm^2.

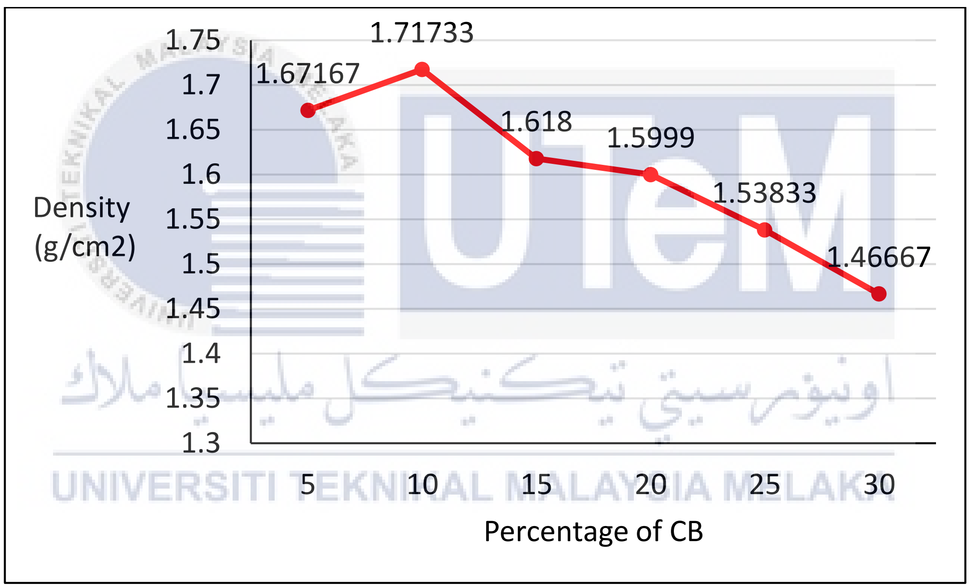


Figure 4.3 Graph of density (g/cm^3) against the percentage of CB

4.4 Hardness

Based on the figure 4.4, a specimen with the percentage of CB 5% has the highest value of hardness which is 60.8 Shore-D. The value of hardness much relies on the content of epoxy as the binder. This is because the value of hardness decreasing linearly with the increasing of CB content. The US-DOE hardness target of the bipolar plate is 40 Shore-D and more. This means that the result meets the US-DOE target.

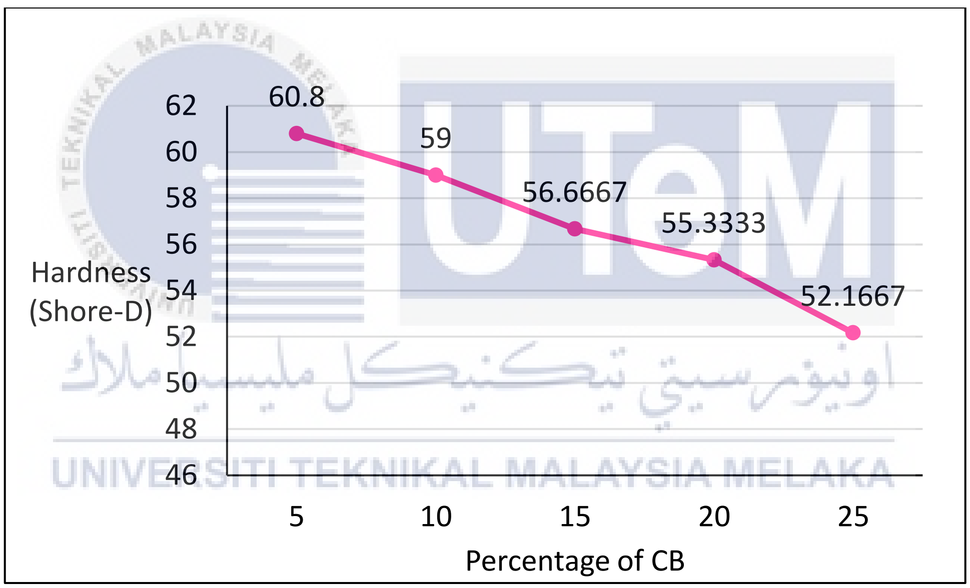


Figure 4.4 Graph of hardness (Shore-D) against the percentage of CB

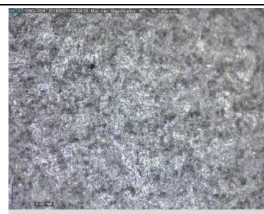
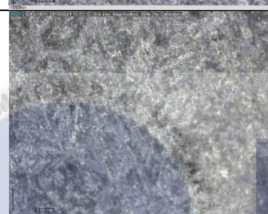
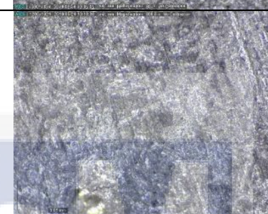
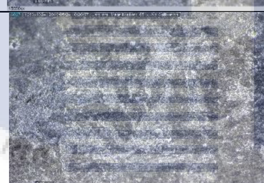
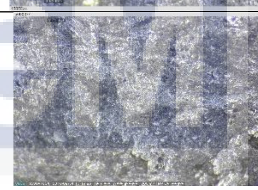
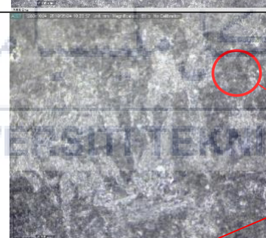
4.5 Microstructure Test

The Figure 4.5 and 4.6 shows the image of Dino Lite (x900) on the surface of different percentage Of CB. It can see that the present of CB is increase when increasing the percentage of CB in the compositions. Table 4.2 shown the microstructure of bipolar plate of different percentage of CB. The bipolar plate with the percentage of CB is over 20% will form a bundle of CB.

Figure 4.5: 5 wt% of CB

Figure 4.6: 25 wt% of CB

Table 4.2: Image of bipolar plate using Dino Lite

Percentage of CB	650x	900x
5		
10		
15		
20		
25		
30		

CHAPTER 5

DISCUSSION

All the four have been carried out and based on it, the result was analyzed and determined to state whether the properties if the G/CB/EP bipolar plate meets the standard set by US-DOE for a bipolar plate. From all the test, only two out of four of the result does not successfully meet the requirements needed by US-DOE which are the electrical conductivity and the flexural strength. While the other two properties are hardness and density manage to meet the requirement.

5.1 Electrical Conductivity Test

There is a maximum limit for carbon black in the composites mixture for the electrical conductivity test. Based on the result obtained, 20% of CB is the maximum loading for the content. The electrical conductivity value of 20% CB is 21.84 S/cm which is the highest value of electric conductivity but it does not meet the requirements set by US-DOE, 100 S/cm. Based on the graph shown in Figure 4.1 the trend of the line is not constant. This can be explained through the article from F. Mighri (2004). This is happening because of the bonding of G and CB particle are not strong enough due to lacking epoxy content.

5.2 Flexural Strength Test

Based on the result obtained from the flexural strength test, the highest value of flexural stress is 10.097 MPa. However, it does not meet the standard requirements set by US-DOE which is higher than 25 MPa. The trend of the line is also not constant or shows a certain pattern. According to F. Mighri (2004), the trend of the line inconstant due to the bonding of G and CB particles are not strong enough because of the lacking of epoxy content.

5.3 Density Test

The density value of all specimen does meet the standard requirement of the US-DOE which is less than 2 g/cm^3. The density test shows that the percentage of CB is inversely proportional with the density value. The maximum density obtained is 1.717 g/cm^3 with the CB content 10%.

5.4 Hardness Test

Based on the result obtained from the hardness test, the maximum value of hardness is 60.8 Shore-D with the CB content 5%. The test shows that the value of hardness is decreased when the percentage of CB increases. The result successfully meets the standard requirement set by US-DOE which is higher than 40 Shore-D.

5.5 Microstructure Test

From the result obtained by using dino lite microscope with the magnification used 600 and 900, the bundle of CB can be seen when the percentage of the CB more than 20% of the total weight. This cause the performance of bipolar plate for the electrical conductivity reduces due to the lack of conductive network between the fillers.

5.6 Effects of Epoxy Content and Mixing Process of Composites

After completely analyzed the result and graph for conductivity and flexural strength, there are several main problems that cause such this result. The main reason that leads to that such trend of flexural strength against the percentage of CB is the amount of epoxy in the compound is not enough to bind the filler content. 20 wt% amount of epoxy is used in this research and it can be seen that the graph is a sudden rise from 5% to 10% content CB content. The reason is the amount of 20% epoxy has not been totally used up to bind the 5% CB content. The graph starts to decrease when the increase of CB because of the epoxy content does not enough to bind the CB content.

The other reason is the process of mixing the carbon black/graphite with the epoxy. The high-speed mixer is used in this research to mix carbon black/graphite with the epoxy. The problems that come about during the mixing process is G/CB/EP is not homogeny mixed up with each other. The time for the mixing process must be taken into consideration. The time to perform the mixing process must not too long because the epoxy will be set in a very short of time. When the compound setting, it cannot be used anymore because it is a thermoset compound where it cannot go through the reversible process.

Those are the two main reason that affects the result of this research and cause the G/CB/EP bipolar plate does meet the standard requirements set by US-DOE for the electrical conductivity and flexural strength. In the future, the other researcher must take consideration of the main problems and perhaps they will find another alternative to mix up the material to get the homogenous mixture.

CHAPTER 6

CONCLUSION AND RECOMMENDATION

6.1 Conclusion

There are two main objectives of this research. First, to study the effect of Carbon Black (CB) on the properties of Graphite (G)-Epoxy (EP) composites. Second, to determine the critical loading of Carbon Black (CB) in G/CB/Epoxy Composites. Therefore, CB content is very important for this research because it can affect the electrical and mechanical properties on the composites. There are several tests was conducted in order to determine the effect of CB in G/CB/EP composites such as electrical conductivity, flexural testing, density, and hardness had been done.

Based on the result obtained from all the research the present of CB content in the compound gives a notable effect on the properties of electrical conductivity, flexural strength, density, and hardness. When the percentage of the CB increases the electrical conductivity does increase too. However, there is a limit for carbon black in the composites mixture for the electrical conductivity test which 20 wt%. Hardness and flexural strength will increase when the CB content in the compound increased but it must have enough amount of epoxy to bind the compound together.

6.2 Recommendations

After all, the test have been carried out and all the result have been analyzing, there are some process or technique that need to improve to achieve the electrical and mechanical properties.

6.2.1 Percentage of the resin content

Filler and resin are two main components that is important to fabricate the composites bipolar plate. The percentage of these components is 80% for filler and 20% for resin. Based on the percolation theory, increasing the amount of filler content will increase the electrical conductivity properties of the composites bipolar plate. It can see that the percentage for the epoxy act as the resin is not enough to hold and bind the filler content. Therefore, for the recommendation, the quantity of the epoxy needs to increase more than 20% of the total weight composition.

6.2.2 Method for the mixing process

In this research, the G/CB/ is mixing with EP by using high-speed blender. This equipment is not the best tools for mixing. When the raw materials not mixed well, it may affect the properties of the bipolar plate composites. Besides, while using the high-speed blender, it produces heat. Therefore, a recommended changing the mixing equipment for binder and filler with better mixing equipment.

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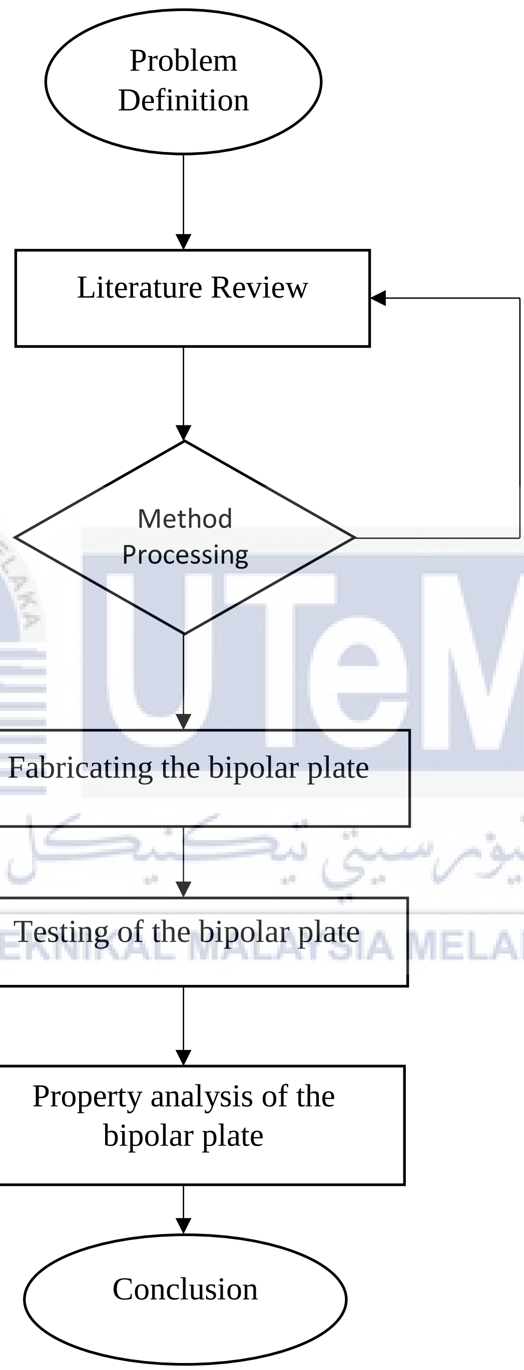
APPENDICES

Flowchart of the research.



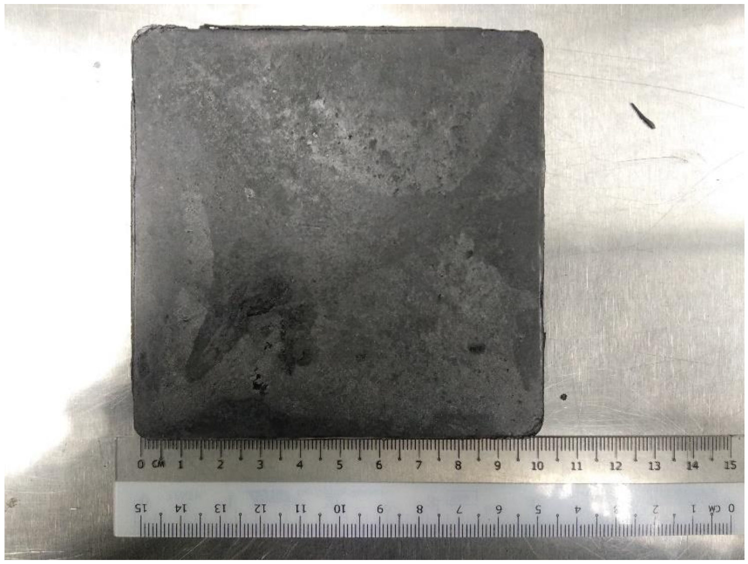
Gantt chart for Final Year Project 1.

Activity	2018			
	September	October	November	December
Selection and confirmation of project title				
Prepare the introduction of FYP 1 1. Objectives 2. Problems statement 3. Scope				
Study the literature review and methodology				
Sample preparation				
Report preparation				

Gantt chart for Final Year Project 2.

Activity	2018				
	January	March	April	May	June
Sample preparation					
Fabrication of bipolar plate					
Testing of bipolar plate					
Properties analysis					
Result and discussion					
Report preparation					

Sample of Specimen

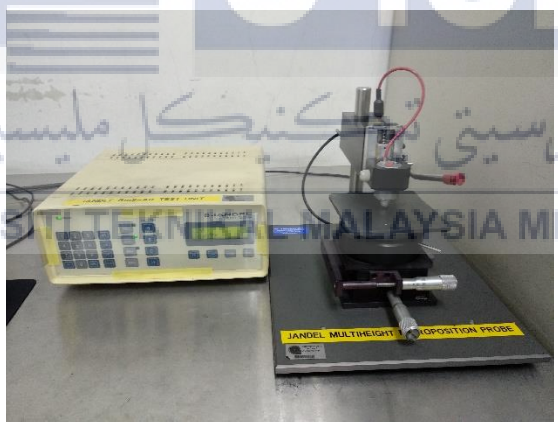


Bipolar plate Sample 100mm x 100mm

Bipolar plate cut to 13mm x 100mm

Test

Flexural strength test



Electrical conductivity test