EFFECT OF CARBON BLACK LOADING ON THE PROPERTIES OF GRAPHITE – EPOXY COMPOSITE FOR FLOW CHANNEL FABRICATION

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DECLARATION

I declare that this project report entitled "Effect of Carbon Black Loading on the Properties of Graphite-Epoxy Composite for Flow Channel Fabrication" is the result of my own work except as cited in the references

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Date : 10/08/2020

APPROVAL

I hereby declare that I have read this project and in my opinion this report is sufficient in terms of scope and quality for the award of the degree of Bachelor of Mechanical Engineering.

Signature : ____________________

Supervisor : ____________________

Date : ____________________

DEDICATION

To my beloved mother and father

ABSTRACT

The addition of conducting fillers into composites will increase the electrical conductivity of the composite. However, the electrical conductivity and other performance aspects of the composite such as flexural strength and hardness will have adverse effect when content of fillers such as carbon black and graphite increased beyond certain extend, which is an effect due to percolation threshold. Conductive fillers are commonly added in polymer composite in fabrication of bipolar plate's component such as flow and cooling channels. The percentage content of each filler material and resin must be in correct proportion to achieve the bipolar plate performance target set up by US DOE. This work studies how the variation of carbon black content in an epoxy–matrix composite which contain another conductive filler: graphite can affect the performance of bipolar plate by examine corresponding variation in electrical conductivity, flexural strength, shore hardness, and bulk density. The flow channels produced are examined by their dimensions and compared with drawing to understand the effectiveness and accuracy of hot compression method in fabrication of flow channel used. This work will provide insight to the best practice of correct proportion to be used in preparing a GR/CB/EP composite for best performance in bipolar plate application and suitability of hot compression method in fabrication of flow channel.

ABSTRAK

Penambahan pengisi konduktif ke dalam komposit akan meningkatkan kekonduksian elektrik komposit. Walaubagaimanapun, kekonduksian elektrik dan aspek prestasi lain komposit seperti kekuatan lenturan dan kekerasan akan merosot apabila kandungan pengisi seperti karbon hitam dan grafit meningkat melebihi komposisi tertentu, disebab kesan ambang perkolasi. Pengisi konduktif biasanya ditambahkan dalam komposit polimer dalam pembuatan komponen plat bipolar seperti saluran aliran dan penyejukan. Kandungan peratusan setiap bahan pengisi dan resin mestilah dalam kadar yang betul untuk mencapai sasaran prestasi plat bipolar yang ditetapkan oleh US DOE. Karya ini mengkaji bagaimana variasi kandungan karbon hitam dalam komposit epoksi-matriks yang mengandungi pengisi konduktif lain: grafit dapat mempengaruhi prestasi plat bipolar dengan memeriksa variasi kekonduksian elektrik, kekuatan lenturan, kekerasan, dan ketumpatan pukal. Saluran aliran yang dihasilkan diperiksa dimensinya dan dibandingkan dengan lukisan untuk memahami keberkesanan dan ketepatan kaedah pemampatan panas dalam pembuatan saluran aliran plat bipolar. Karya ini akan memberi gambaran mengenai amalan terbaik komposisi yang betul untuk digunakan dalam penyediaan komposit GR / CB / EP untuk prestasi terbaik dalam aplikasi plat bipolar dan kesesuaian kaedah pemampatan panas dalam pembuatan saluran aliran.

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LIST OF ABBREVIATIONS

Abbreviation	**Full Name**
BP	Bipolar Plate
CB	Carbon Black
CMM	Coordinate Measuring Machine
CPC	Conductive Polymer Composite
EP	Epoxy
GCC	Ground Calcium Carbonate
GDL	Gas-Diffusion Layer
Gr	Graphite
MEA	Membrane Electrode Assembly
PCC	Precipitated Calcium Carbonate
PEMFC	Proton-Exchanged Membrane Fuel Cell
US DOE	United States Department of Environment

CHAPTER 1

ITRODUCTION

1.1 BACKGROUND

Conductive polymer is a composite material which added conductive fillers into polymer. It can conduct electricity but poorer than metal and pure graphite but can work in high corrosive environment which is something that metal cannot achieve. The metal bipolar plate requires extra coating process on its surface to provide corrosion resistance while pure graphite bipolar plate requires resin impregnation to fill up the porous holes and avoid water permeation, these extra processes are costly when mass production in commercial purpose which resulted in the surge of conductive polymer composite (CPC) as the alternative for the bipolar plate material. CPC is largely used as a material to fabricate bipolar plate in fuel cell due to its sufficient electrical conductivity, light-weight, low cost and corrosion resistance. Besides, the high formability also eases the fabrication of flow field design on the bipolar plate.

Most of the researches at the field of CPC up to date have added conductive fillers such as carbon black, carbon nanotube, graphite, metal powders into polymer composites to increase electrical conductivity. Out of all, carbon black is most commonly added into the graphite-based polymer composite due to it can increase the mechanical strength and its high specific surface area can form a conductive network which greatly increase conductivity in plastics even in small amounts. Current researches conducted are focused on how to use the cheapest material to form the most extensive network in the matrix or in other word decrease the percolation threshold (Hyun-Jung Choi, et al, 2019).

Bipolar plate is the key component of proton-exchanged membrane fuel cell (PEMFC). Its functions are distributing gas and fuel evenly, remove heat from the active area, conduct electricity from cell to cell, and act as sealant of fuel cell. It represents about 80% of the total weight and 30 to 40% of cost of fuel cell. Hence, it is a great challenge to found materials which possess the mechanical properties and processibility of polymer as well as good conductivity of metal (E. Planes, et al, 2012). These completely different properties are not present at singular material, hence underscore the need of composite material which incorporated all the advantages of different materials.

The US DOE has set up technical targets to achieve for the bipolar plate fabricated for transportation uses. The targets are listed at Table 1.1. Few of them will be served as the benchmark for this study's fabricated bipolar plate samples.

Table 1.1 Technical Targets for Bipolar Plate for Transportation Application (Fuel Cell Technologies Office, 2019)

Characteristic	2015	2020
Cost (\$/kW_{net})	7	3
Plate Weight (kg/kW_{net})	<0.4	0.4
Plate HH_2 permeation coefficient (Std cm^3/(sec cm^2 Pa) @ 80°C, 3 atm, 100% RH)	0	<1.3 x 10^{-14}
Corrosion, anode (µA/ cm^2)	no active peak	<1 and no active peak
Corrosion, cathode (µA/ cm^2)	<0.1	<1
Electrical conductivity (S/cm)	>100	>100
Areal specific resistance (ohm cm^2)	0.006	<0.01
Flexural strength (MPa)	>34 (carbon plate)	
Forming Elongation (%)	20-40	40

1.2 PROBLEM STATEMENT

Fuel cell is one of the greenest fuels at the world since its waste product is only water. PEMFC is predicted as the future of the fuel source used in automobile. The production and shipping of PEMFC is increasing yearly mainly due to transportation uses. However, there are still plenty of room for improvement in terms of cell lifetime, tolerance for impurities at electrode, and design optimization.

Bipolar plates which is one of the key components of PEMFC need to provide electrical conduction between the cells, high resistant to chemical corrosion and has very low gas permeability. These requirements are steep challenges for any class of materials, none of them can fits the needs totally. This has leaded to the only resolution: invention of new material. Many researches have been conducted to find out appropriate materials to fabricate bipolar plate.

Up to date, there are several options which have potential to be materials for bipolar plates to realize target of commercialization of PEMFC which are metal with or without coating, non-porous graphite and graphite based polymer composite. The weakest point of polymer composite is low electrical conductivity when compared to metal and graphite materials although it is lower in weight, cost and ease in fabrication. Hence, the current challenge faced by scientists around the world is to invent composites with better electrical properties at low cost. Various types of conductive fillers such as graphite, carbon nanotubes, carbon black and metallic powders are employed to increase the conductivity. The percentage of fillers and binder in a composite are investigated in many researches to find out the most optimum parameters for various requirements as stipulated in US DOE targets for bipolar plate in transportation application at lowest cost.

The flow field design and flow channel dimensions also affect the local current density, temperature and water content of PEMFC (S. Shimpalee, 2011). Proper design of the dimensions of flow channel can avoid flooding at electrode and dehydration of MEA which affect the performance of PEMFCs significantly.

There are various fabrication processes for flow channel such as machining and hot compression method. Machining method made use of CNC machine to cut out a high accuracy dimension of flow channel at graphite plate but is time consuming and high cost. In contrast, hot compression method is easy and cheaper to employ in fabrication of flow channel, but the dimension accuracy is inferior to machining method.

1.3 OBJECTIVES

1. Study the effect of Carbon Black on the properties of Graphite/Epoxy composite.
2. Determine the parameter dimension of flow channel.

1.4 SCOPE OF PROJECT

The effect of carbon black on the properties of graphite/epoxy composite will be determined using 3 different weight percentages of carbon black and graphite fillers with fixed weight percentage of epoxy as binder. This composite will be fabricated through hot compression molding. During fabrication process the flow and cooling channel will be fabricated on the surface of the sample. The properties determined from mechanical and electrical tests will be analyzed to get the critical loading of carbon black in the composite. The dimension of flow channel will be determined after fabrication of bipolar plates and compared with mold to analyze the difference in dimension.

CHAPTER 2

LITERATURE REVIEW

2.1 FUEL CELL

39 years after the invention of the world first battery, fuel cell is invented which its biggest difference with the previous is it acts as a device to convert external fuel source, hydrogen to react with oxygen where the chemical process generates electricity, instead of storing energy. Fuel cell generates electricity in a simple step through chemical reaction when compared with existing power plant which undergone many processes to generate electricity. This enable fuel cell as future energy source due to its clean, green and efficient characteristics.

Fuel cell can be varying in the forms of different electrolytes which are potassium hydroxide solution (alkali fuel cell), metal carbonates salt (molten carbonate fuel cell), phosphoric acid (phosphoric acid fuel cell), polymer sheet (proton exchange membrane fuel cell) and metal ceramic compound (solid oxide fuel cell). The first three of the electrolytes are in liquid form while the last two are in solid form.

All kinds of fuel cell operated in slightly different conditions but the process can be summarized as hydrogen oxidized at anode and reduced by oxygen at cathode to form water, the emission of electron during oxidation process will induced a DC current flow through external circuit.

2.2 POLYMER ELECTROLYTE MEMBRANE FUEL CELL (PEMFC)

PEMFC has an anode which acts as negative terminal and cathode acts as positive terminal of the cell. At anode, hydrogen molecules undergo oxidation to eliminate electrons to become hydrogen ion. The electrons emitted are conducted through external circuit. The ionized hydrogen is then reduced to water by oxygen at cathode. The half chemical equations are:

$$H^2 - 2e^- = 2H^+ \text{ (Anode)}$$

$$O_2 + 4e^- + 4H^+ = 2H_2O \text{ (Cathode)}$$

The schematic diagram of the working order for a single PEMFC is shown in Figure 2.1.

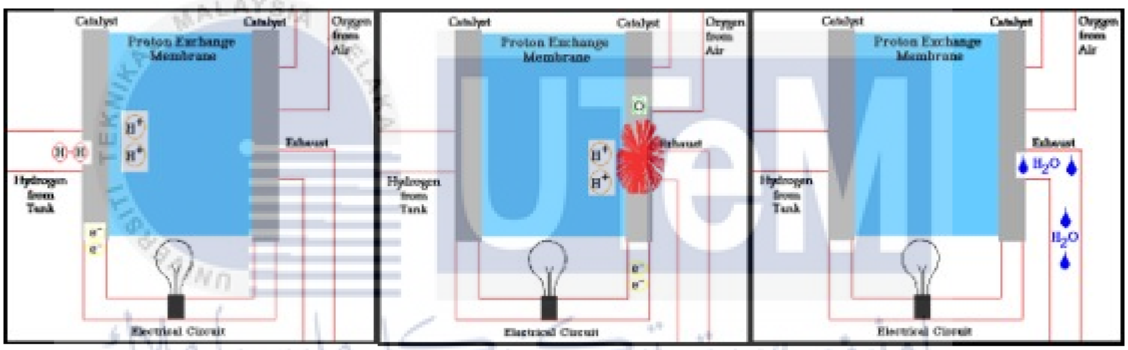

Figure 2.1 Single PEMFC (Smithsonian, 2017)

There is a channel connected to each terminal of the cell to provide hydrogen and oxygen supply to anode and cathode respectively. Since both of the electrodes at the terminals are made up of precious metal and has low operating temperature (< 100 °C), pure fuel and oxygen sources are needed. The electrolytes used in PEMFC is ion-selective membrane made up of ionomers. The combination of ionomeric membrane sandwiched between two electrodes thin films is known as membrane electrode assembly (MEA) which is the key component of PEMFC.

There are two bipolar plates at the end of single fuel cell which usually made up of high density graphite. There are gas flow field machined on the plates to allow even distribution of react gases with electrodes which is further aided by presence of gaskets which helps limit the flow of gases on the effective area and gas-diffusion layers (GDL) which enable direct access of the gases with catalysts. PEMFC is stacked to provide voltage according to demand. The current is conducted from one cell to another through the bipolar plates. The main components of single PEMFC is shown in Figure 2.2. Since the PEMFCs is dynamic, light weight and able to operate under low temperature, it is usually used in vehicle.

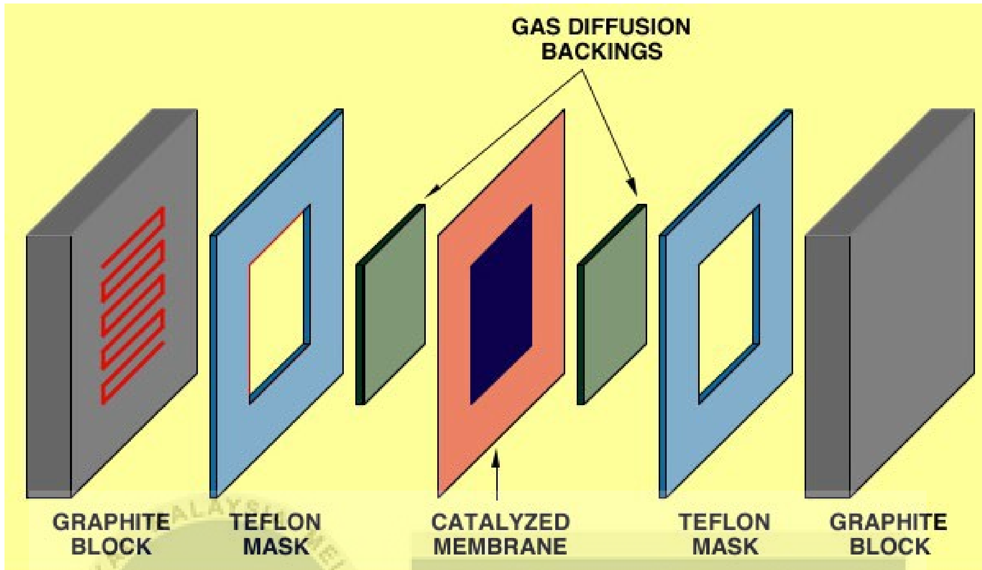

Figure 2.2 Main Components of Single PEMFC (Gottesfeld, n.d.)

2.3 FLOW CHANNEL

A flow channel is the channel on bipolar plates which helps to provide adequate amount of react gases (hydrogen and oxygen) to gas diffusion layer (GDL) and surface of the catalysts meanwhile maintain the flow pressure. It comes with different often-used shapes as shown in Figure 2.3.

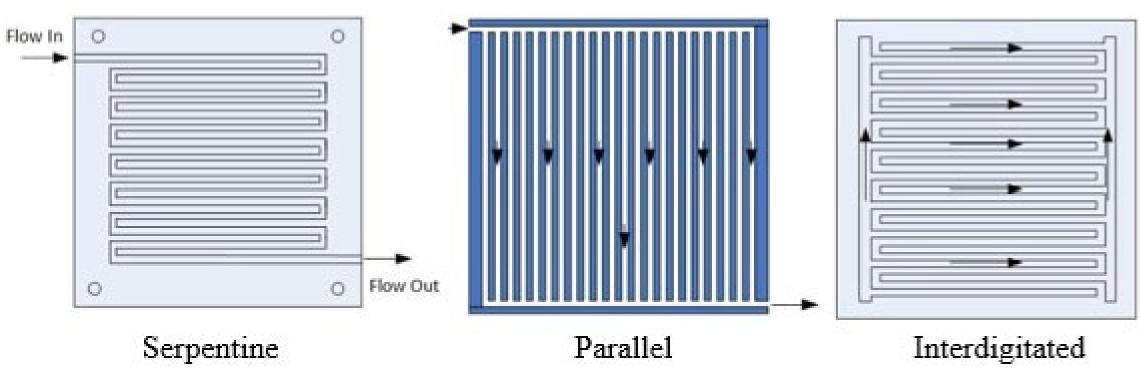

Figure 2.3 Flow Channel Shapes (Spiegel, 2017)

Serpentine flow field design is relatively efficient at providing flow distribution across the electrode surface of the fuel cell. This design may cause pressure loss due to the relatively long flow path (Spiegel, 2017). The most qualitative flow field for PEMFC applications proved to be the serpentine flow field, having a higher pressure drop than parallel flow field and efficiently removing accumulated water from the channels (Ionescu, 2018).

Flow channel dimensions also affect the performance of PEMFC. To achieve the highest power density in a PEM fuel cell, it requires that both the total width and the rib ratio be small. In the small total width, the flow channel width is small and water can easily diffuse from the corner to the middle of the channel (in cross-section) to hydrate the membrane, which increases the membrane conductivity and thus the cell performance (Liu, 2013). According to Perng, S.-W., & Wu, H.-W. (2015), the larger angle and height of trapezoid baffles significantly strengthen the blockage effect and thus promote the cell performance. However, the baffles of 1.5 mm and 90° cause local high temperature (>372 K) to damage the membrane of PEMFC and the maximum pressure drop. As a result, the maximum enhancement of cell net power from a novel PEMFC with baffles of 60° and 1.125 mm is about 90%.

2.4 CONDUCTIVE POLYMER COMPOSITE (CPC)

Bipolar plates (BP) is one of the key components of PEMFC which transport react gases, water and conduct electron. According to Ehsan Zarmehri, et al. (2013), BP has the most significant role in weight, volume and corresponding costs of fuel cells. In order to be used in automotive applications, BP must be light weight and small in volume, have shock resistance and can withstand harsh operating environment. A lot of researches has been done to find out better materials to build BP which range from metals to composites, which are more cost effective and feasible than traditionally used high-density graphite. This is due to pure graphite is brittle, porous and complicated in machining flow field (Heinzel et al., 2009).

Conductive polymer composite (CPC) is graphite composite material which is used to fabricate BP used in static PEMFC applications. It utilizes the combination of conductive fillers such as carbon nanotubes, carbon blacks, graphene and binder such as epoxy and phenolic resins, polyester, polypropylene to provide low cost, light weight, high conductivity, process ability, corrosion resistance and mechanical strength properties. According to Heinzel et al. (2009), the conducting fillers form a percolation network within the polymer matrix with a required good quality of dispersion via a kneader or extruder, and usually incorporated with graphite fibers to increase mechanical strength.

2.5 MATERIALS

Materials used in production of BP in this project are CPC, made up of Gr and CB as fillers while EP as binder. The physical properties of the materials used to produce BP samples are listed in the Table 2.1.

Table 2.1 Physical Properties of Gr/CB/EP

Properties	Gr	CB	EP
Density (g/cm^3)	2.09 - 2.23	1.8 - 2.1	1.54
Electricity Conductivity at 20°C (S/m)	3.3×10^2	-	-
Volume Resistivity (ohm-cm)	-	-	$> 10^{10}$
Thermal Conductivity (W/mK)	< 1950	-	1.30 - 2.88
Flexural Strength (MPa)	15.5 - 207	-	1.12×10^8

2.5.1 Filler

Filler can increase specific properties and reduce the cost of composite. The top filler materials used are ground calcium carbonate (GCC), precipitated calcium carbonate (PCC), kaolin, talc, and carbon black (Market Study: Fillers (3rd edition), 2014). Fillers mainly used to fill up space in targeted area, increase material strength, conductivity and surface smoothness.

2.5.1.1 Carbon Black (CB)

Carbon black is the generic name for a family of small-size, mostly amorphous, or para crystalline carbon particles grown together to form aggregates of different sizes and shapes, primary particle size, chemistry, porosity and surface area according to commercial grades. CB is formed in the gas phase by the thermal decomposition of hydrocarbons in the absence or presence of oxygen in sub stoichiometric (Michael E. Spahr, Roger Rothon, 2016).

According E.B. Sebok, et al. (2001), ~93% of CB usage is in rubber applications, and can be classified into two main categories: tires and mechanical rubber goods (e.g. automotive belts and hoses). The reinforcing properties of carbon black provide enhanced performance and durability to the rubber compounds in which they are used.

CB also used in graphite-polymer composite bipolar plate due to its structural characteristics such as morphology and size have decisive influence on the final properties of bipolar plates. The small carbon black particles helped to make conducting tunnels between graphite particles, increasing conductivity (Renato et al., 2011).

2.5.1.2 Graphite (Gr)

Graphite (Gr) is a crystalline form of the element carbon with its atoms arranged in a hexagonal structure. This structure enables Gr has high degree of anisotropy which determine its properties. It is extremely soft, cleaves with very light pressure, and has a very low specific gravity. In contrast, it is extremely resistant to heat and nearly inert in contact with almost any other material (King, n.d.). According Isabel Suárez-Ruiz, et al (2008), Gr shows a high directional strength and is highly inert, although its reactivity increases as the temperature rises and therefore it reacts with oxygen at temperatures higher than 300–400°C.

Gr is a common material used to make bipolar plate. The corrosion for graphite-base composite is negligible compared with metallic bipolar plate. It offers advantages in weight, formability and cost. However, it has lower strength and electrical conductivity than steel plates.

Composite plates in current field use are thicker than metal plates, resulting in larger stacks. Newer state-of-the-art composite bipolar plates are thinner and approaching the thickness of metal plates but the long curing time for resins extend the processing time (John P. Kopasz, Thomas G. Benjamin, 2017).

2.5.2 Binder

Binder is used to hold the strengthening material together to form a composite. It protects the fibers from external and environmental damage and transfer the load between the fibers. Materials of binder can be organic such as thermoset, thermoplastic and rubber as well as inorganic like cement, metals and ceramic.

2.5.2.1 Epoxy (EP)

Epoxy resins are a family of monomelic or oligomeric material that can be further reacted to form thermoset polymers possessing a high degree of chemical and solvent resistance, outstanding adhesion to a broad range of substrates and a low order of shrinkage on cure (C. Craver, C. Carraher, 2000). It is widely used as matrix of polymer composite, repairing of concretes and structures, manufacture of adhesives, flooring, plastics, sealers, paints and coatings. It has strong mechanical properties and can work over temperature of -250°C to 175°C.

2.6 MANUFACTURING PROCESS

There are two main fabricating methods for bipolar plate, which are injection molding and compression molding. Both processes come with pros and cons, hence the best approach is depending on the requirement of output volume, product geometry and accuracy, cost and accessibility. The fabrication methods are shown in Figure 2.4.

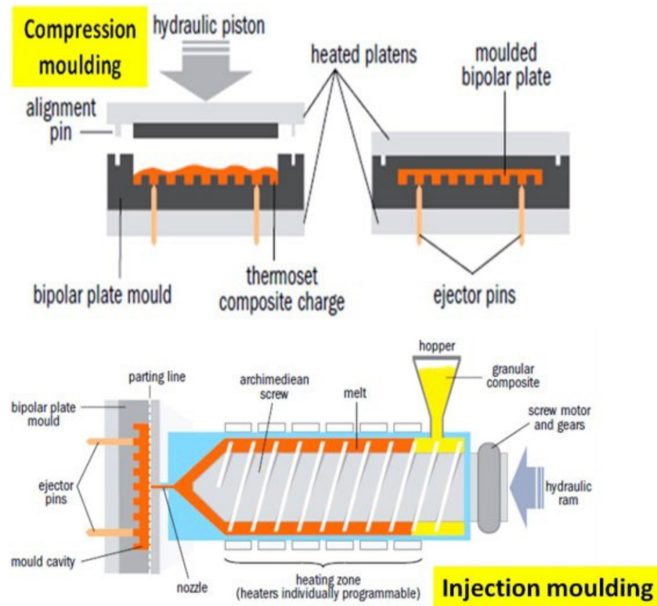

Figure 2.4 Fabrication Methods of Bipolar Plate (Rungsima Yeetsorn et al., 2011)

2.6.1 Injection Molding

Injection molding is the process where material is heated then injected into a mold cavity. This is the perfect molding method for complex shapes. It also is great for easily changing sizes and uptime efficiencies due to independent cavity control. Injection molding is usually the method of choice for a lower volume run because of its easy line changes and multiple material choices. The disadvantages of injection molding are longer color changes, large space footprint and gates present can cause post-printing challenges (Yonkers, 2018). Injection molding used in production of bipolar plate due to very short cycle times hence very cheap. According Heinzel et al. (2009), it is mostly used with thermoplastic resin.

2.6.2 Compression Molding

Compression molding is a process of pouring thermosetting resins into a heated mold cavity and then forced down with another implement. As the material is distributed through the mold, it is kept at a preset temperature and pressure level until it has cured. Typically, compression molding is used for applications that require high-volume output (KASO, 2018). It also has lower maintenance cost for required equipment's, less material waste and low energy cost. The disadvantages of compression molding are longer changeover, difficult for complex part geometry and often less consistent than injection molding (Yonkers, 2018). Compression molding is suitable for both thermoplastic and thermoset binders but is more cost-effective with the latter due to shorter cycle times.

2.7 TESTING METHOD

The fabricated bipolar plates with flow channel will undergo several types of tests to determine its electrical and mechanical properties. The common electrical property of bipolar plate to be determined is electrical conductivity while for mechanical properties, the bipolar plate shall be tested its flexural strength, bulk density and shore hardness.

2.7.1 Electrical Property

Electrical conductivity is the most common electrical property to evaluate the performance of bipolar plate. Conductivity is defined as the ratio of the density of current to the strength of the electric field. The electrical conductivity can be calculated measuring the resistance, area and length of the test material. A four-terminal ohmmeter is used for greater accuracy. This type of ohmmeter is more accurate because one pair of terminals measures current, while the other pair measures voltage. This allows the ohmmeter to ignore the resistance of the first pair of terminals (Robinson, 2017).

2.7.2 Mechanical Properties

The first mechanical properties of bipolar plate to be determined is flexural strength. Flexural strength is defined as the maximum stress at the outermost fiber on either the compression or tension side of the specimen. The flexural properties of a specimen are the total effect of all three stresses: compressive, shear and tensile as well as the load applied and is less affected by the its geometry. The test can be completed with the 3 and 4-point flexural bend test equipment (Flexural Test, 2019).

The second test is shore hardness which is a measure of the resistance a material has to indentation. Hardness of polymer materials such as Thermoplastics and Thermosets are measured by Shore D scale. Test is done with durometer which utilizes an indenter loaded by a calibrated spring. Hardness is determined by the penetration depth of the indenter under the load (Kopeliovich, 2013).

Bulk density is another important data to be collected as a performance indicator of bipolar plate as it determines the weight of bipolar plate. It is defined as the weight of dry powder divided by the known powder volume. The old method of estimating bulk density of fibers is by placing a known quantity of fiber into a graduated syringe and applying sufficient pressure to pack the content in the syringe while recording the final volume (Parrott et al., 1978). Current method has simplified the measurement by using digital bulk density meter.

2.7.3 Dimension Property

The dimension of flow channel will be measured with Coordinate Measuring Machine in Coordinate Measurement Test. The machine is capable to measure the geometry of an object by sensing discrete points on the surface of the object with a probe. It allows probe move along the X, Y, and Z axes and some devices' probe angle can be control to reach part surface that is otherwise unreachable.

CHAPTER 3

METHODOLOGY

3.1 EXPERIMENT OVERVIEW

The experiment will undergo several processes. Firstly, the preparation of raw materials according to planned composite composition. Next, the pre-mixing and mixing process which mix different composite materials together. Subsequently, the fabrication of sample which include flow channel. Lastly, the sample produced will undergone several mechanical and electrical conductivity tests as well as dimension accuracy test for the flow field to collect data result for further analysis.

3.2 COMPOSITE COMPOSITION

The material used to fabricate composite are conductive fillers which are Gr and CB, and EP as polymer binder. The total weight percentage of fillers is fixed at 75 % while for binder is fixed at 25% due to recent researches showed that at this ratio will produce bipolar plate with most optimize compromise between electrical conductivity and mechanical properties. The fabrication temperature for the composite will be set at 100 °C as at this temperature will produce bipolar plate with relatively high electrical conductivity. There will be three samples with varied weight percentages of Gr and CB fabricated as shown in Table 3.1.

Table 3.1 Composition of Gr/CB/EP Composite

Gr (wt%)	CB (wt%)	EP (wt%)	Molding Temperature (°C)
35	40	25	100
40	35	25	100
45	30	25	100

3.3 FLOW CHANNEL CHARACTERISTICS

Serpentine flow field will be selected to be the design of the sample flow channel fabricated and the clearing is in V-shape as the shape is most suitable to be fabricated by hot compression molding method which will be utilized in the experiment. The dimension of the center part mold is shown in Figure 3.1.

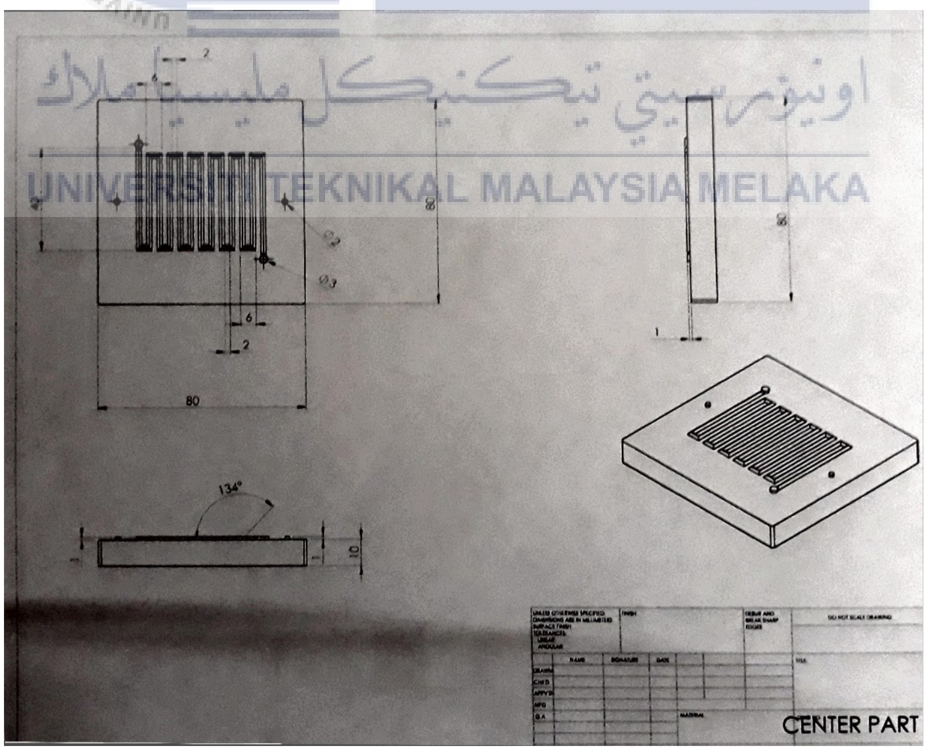

Figure 3.1 CAD Drawing for Center Part Mold (M.Z. Selamat, 2019)

3.4 FABRICATION METHOD

After the preparation of different composition of composite raw materials, the fabrication process will be taking place from the first step, pre-mixing using ball mill machine to mix the different fillers together. In the subsequent internal mixing process, the mixed fillers will be mix with binder before put into hot compression mold to fabricate BP samples.

3.4.1 Pre-Mixing

Pre-mixing of fibers is crucial to fabricate a composite with evenly distributed fiber types and maintain same mechanical and electrical properties at each point of the composite fabricated. The raw materials which are in powder form will be weighted according to planned ratio and the total weight of Gr and CB fibers for each sample is 30 g as shown in Table 3.2. The weighted fibers powder will be poured into the ball mill machine as shown in Figure 3.2. The pre-mixing process will be left to run half an hour to ensure thoroughly mixed of different fibers.

Table 3.2 Composition and Weight of Gr and CB Fibers

Gr			**CB**		
wt%	**%**	**gram**	**wt%**	**%**	**gram**
45	60.00	18	30	40.00	12
40	53.33	16	35	46.67	14
35	46.67	14	40	53.33	16

Figure 3.2 Ball Mill

3.4.2 Mixing

The mixed fillers, Gr/CB will be mixed with polymer matrix, EP by using blender as shown in Figure 3.3. The ratio of filler to binder is 3:1. The process is to ensure fillers is well dispersed and randomly distributed in polymer matrix. The total weight of epoxy and hardener is 10 g for each sample, in ratio of 3: 2, which are 6 g of epoxy and 4 g of hardener.

Figure 3.3 Blender

3.4.3 Compression Molding

Compression molding is the method used in the experiment to fabricate the flow channel samples. The mold with Serpentine flow field and V-shape clearing as shown in Figure 3.4 is pressed onto the powder by using Hot Press Machine as shown in Figure 3.5. All the specimens are undergoing preheating of 15 minutes at pressure of 10 tons before compressed at pressure of 100 tons over 15 minutes, at a temperature set at 100 ℃ at both stages.

Figure 3.4 Mold

Figure 3.5 Hot Press Machine

3.5 TESTING METHOD

The fabricated flow channel samples will have undergone several mechanical, electrical and dimension accuracy tests to investigate the composition of fillers towards performance of bipolar plate and accuracy of parameter dimension of flow channel compared to mold under hot compression fabrication method.

3.5.1 Electrical Conductivity

The electrical conductivity of the samples fabricated are measured by using Jandel Multi Four Point Probe. The device can measure the resistivity of specimen by forcing a constant current across the outer 2 probes to obtain reading of resultant voltage across the inner 2 probes. The device is shown in Figure 3.6.

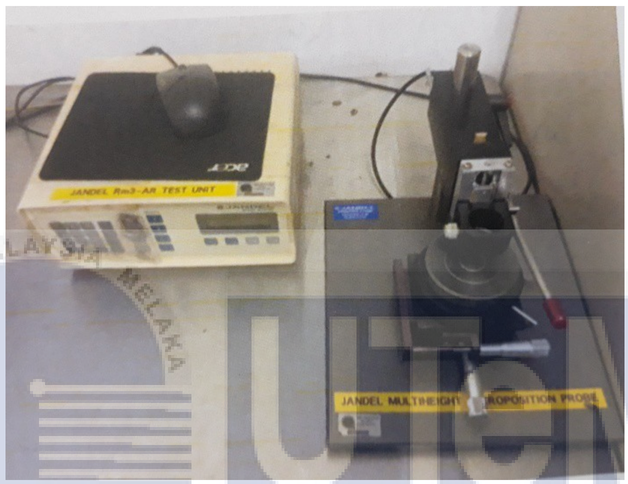

Figure 3.6 Jandel Multi 4 Point Probe

3.5.2 Flexural Strength

The flexural strength of the samples fabricated are measured by using Instron Universal Testing Machine as shown in Figure 3.7. The method used is three-point flexure test method. The specimen is centrally loaded between 2 supports below the specimen. The flexural response of the specimen is obtained by recording the load applied and the resulting strain.

Figure 3.7 Instron Universal Testing Machine

3.5.3 Shore Hardness

The shore hardness of the samples fabricated will be tested using durometer which is based on ASTM D2240 Type D scale as shown in Figure 3.8. The pressure foot of durometer will apply load on the specimen which causing an indentation on the surface of specimen. The depth of the indentation implied the hardness of the specimen tested.

Figure 3.8 Teclock GS-702G Durometer

3.5.4 Bulk Density

The bulk density of bipolar plate samples fabricated are measured by using Electronic Densimeter as shown in Figure 3.9. The sample will first be placed on the device's container to be weighted, and then put into the liquid medium to determined its volume and thus the derivate bulk density.

Figure 3.9 Electronic Densimeter

3.5.5 Coordinate Measurement

The dimension of flow channel is measured by using the Coordinate Measuring Machine smart scope CNC500. The device measures the length of a physical object by sensing discrete points on the surface of an object with a probe. An example of Coordinate Measuring Machine is shown in Figure 3.11.

Figure 3.11 Coordinate Measuring Machine [Photograph of Axiom too 1200 DCC CMM] (2019). Retrieved from https://www.amazon.com/Axiom-too-1200-DCC-CMM/dp/B07JFR4M3Y

CHAPTER 4

DATA AND RESULT

4.1 SAMPLES OBSERVATION

The samples are fabricated using Hot Press Machine. Figure 4.1 showed the samples in mixed powder form which is prepared after pre-mixing process. Table 4.1 showed the parameters used in preparing the samples when using Hot Press Machine.

Figure 4.1 Samples in Powder Form after Pre-Mixing

Table 4.1 Parameters Used in Preparing Samples when Using Hot Press Machine

Sample	Composite Composition (GR/CB/EP)	Types of Channel	Composite Mixing Time (Min)	Pre-heating Duration (Min)	Pre-heating Pressure (Tons)	Compression Duration (Min)	Compression Pressure (Tons)	Pre-heating & Compression Temperature (°C)
1	45/30/25	Flow Channel	5	15	10	15	100	100
2	45/30/25	Cooling Channel	5	15	10	15	100	100
3	40/35/25	Flow Channel	5	15	10	15	100	100
4	40/35/25	Cooling Channel	5	15	10	15	100	100
5	35/40/25	Flow Channel	5	15	10	15	100	100
6	35/40/25	Cooling Channel	5	15	10	15	100	100

Table 4.2 Samples of Gr/CB/EP

Samples / Gr/CB/EP	Flow Channel	Cooling Channel
45/30/25	 Sample 1	 Sample 2
40/35/25	 Sample 3	 Sample 4
35/40/25	 Sample 5	 Sample 6

4.2 ELECTRICAL CONDUCTIVITY TEST

The electrical conductivity of samples 2, 4 and 6 which represents 3 different compositions are tested on 9 equally divided areas for each of them in both front and back orientation as shown in Figure 4.2.

Figure 4.2 Locations of Testing Site of Samples for Electrical Conductivity Test

The results of electrical conductivity for samples 2, 4 and 6 are tabulated in Table 4.3. and shown as graph in Figure 4.3.

Table 4.3 Electrical Conductivity of Samples at 3 Different Composition

Sample	CB (wt%)	Electrical Conductivity, σ (S/cm)
2	30	54.59
4	35	64.27
6	40	70.03

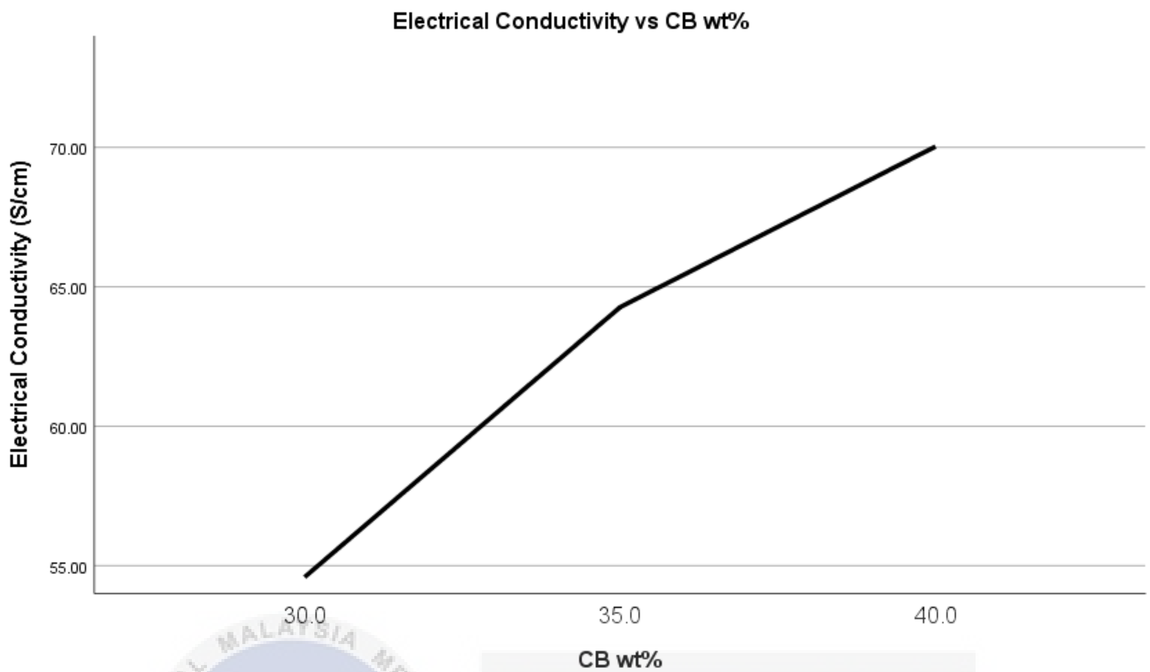

Figure 4.3 Graph of Electrical Conductivity vs CB wt%

From Table 4.3, the electrical conductivity increase 17.7 % from 54.59 S/cm to 64.27 S/cm when CB content increase from 30 to 35 wt%. When CB content increase another 5 wt% to 40 wt%, the electrical conductivity only increase 8.9 % to 70.03 S/cm. The graph in Figure 4.3 showed that electrical conductivity increased when weight percentage of CB increased but the magnitude of increment getting smaller each times.

4.3 FLEXURAL STRENGTH TEST

The flexural strength of the 3 different composition samples is measured on Samples 2, 4 and 6 cooling channels using three-point method. Each testing sample is cut into a rectangular specimen with dimensions of length 100 mm, width 13 mm and thickness of 3 mm. The span length at both left and right are 25 mm. The tests were conducted at room temperature by programmed Instron Universal Testing Machine at 50 kN load and ramp rate of 0.1 mm/s.

Table 4.4 Flexural Strength of Samples at 3 Different Composition

Sample	CB (wt%)	Flexural Strength (MPa)
2	30	2.71
4	35	4.55
6	40	3.89

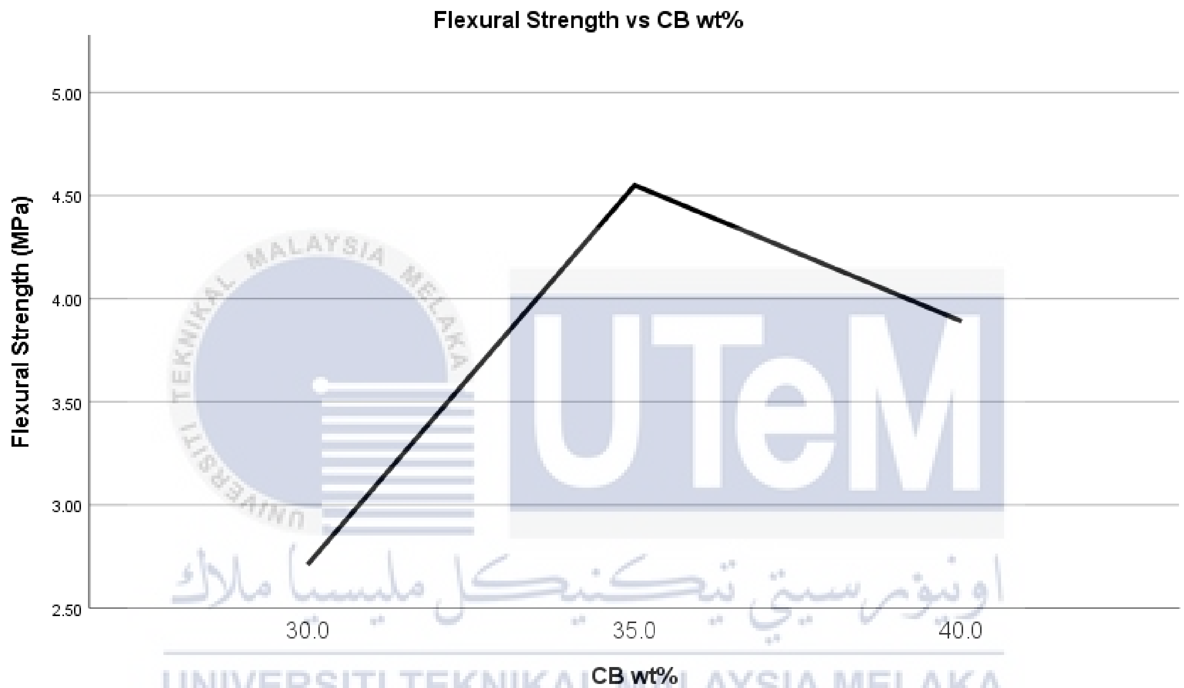

Figure 4.4 Graph of Flexural Strength vs CB wt%

From Table 4.4, the flexural strength increase 167.9 % from 2.71 MPa to 4.55 MPa when CB content increase from 30 to 35 wt%. When CB content increase another 5 wt% to 40 wt%, the flexural strength decrease 14.5 % to 3.89 MPa. The graph in Figure 4.4 showed that flexural strength increased when weight percentage of CB increased until 35 wt% and further increase in CB weight percentage will decrease the flexural strength.

4.4 HARDNESS TEST

The shore hardness of the 3 different compositions samples is measured on Samples 2, 4 and 6 cooling channels. From Table 4.5, the shore hardness increase 17.6 % from 25.65 to 30.16 when CB content increase from 30 to 35 wt%. When CB content increase another 5 wt% to 40 wt%, the shore hardness further increase 20.7 % to 36.41. The graph in Figure 4.5 showed that shore hardness increased when weight percentage of CB increase and the magnitude of increments are slightly increased after each addition of CB weight percentage.

Table 4.5 Shore Hardness of Samples at 3 Different Composition

Sample	CB (wt%)	Shore Hardness
2	30	25.65
4	35	30.16
6	40	36.41

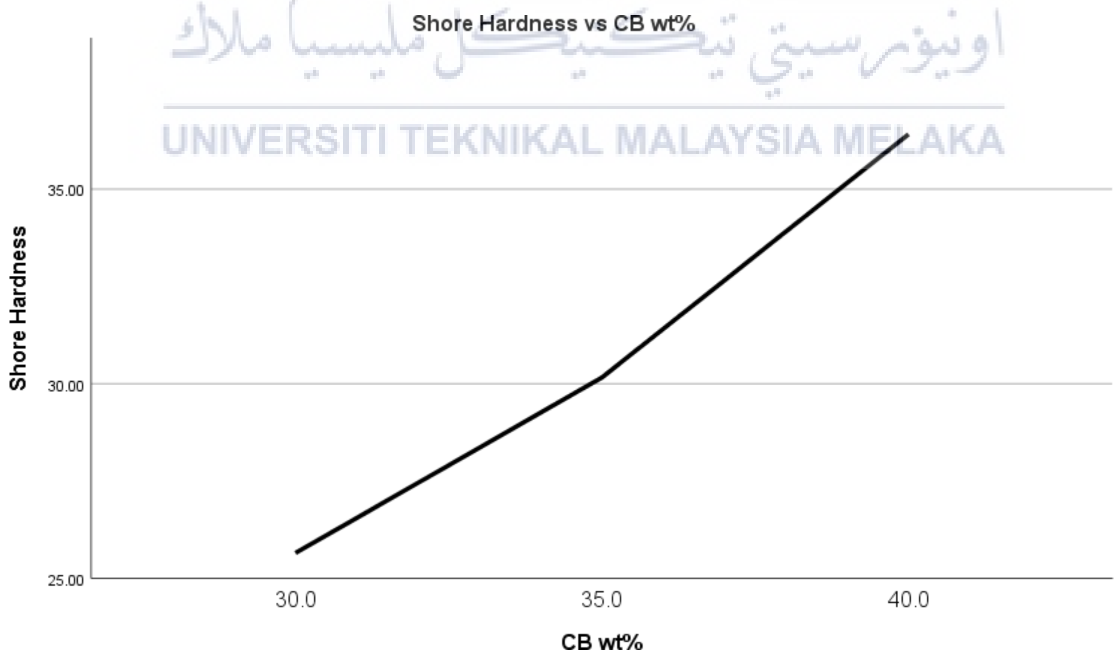

Figure 4.5 Graph of Shore Hardness vs CB wt%

4.5 BULK DENSITY TEST

The bulk density of the 3 different compositions samples is measured on Samples 2, 4 and 6 cooling channels. From Table 4.6, the bulk density decrease 6 % from 1.763 g/cm^3 to 1.658 g/cm^3 when CB content increase from 30 to 35 wt%. When CB content increase another 5 wt% to 40 wt%, the shore hardness further decrease 5.1 % to 1.573 g/cm^3. The graph in Figure 4.6 showed that bulk density almost decreased linearly with CB weight percentage.

Table 4.6 Bulk Density of Samples at 3 Different Composition

Sample	CB (wt%)	Density (g/cm^3)
2	30	1.763
4	35	1.658
6	40	1.573

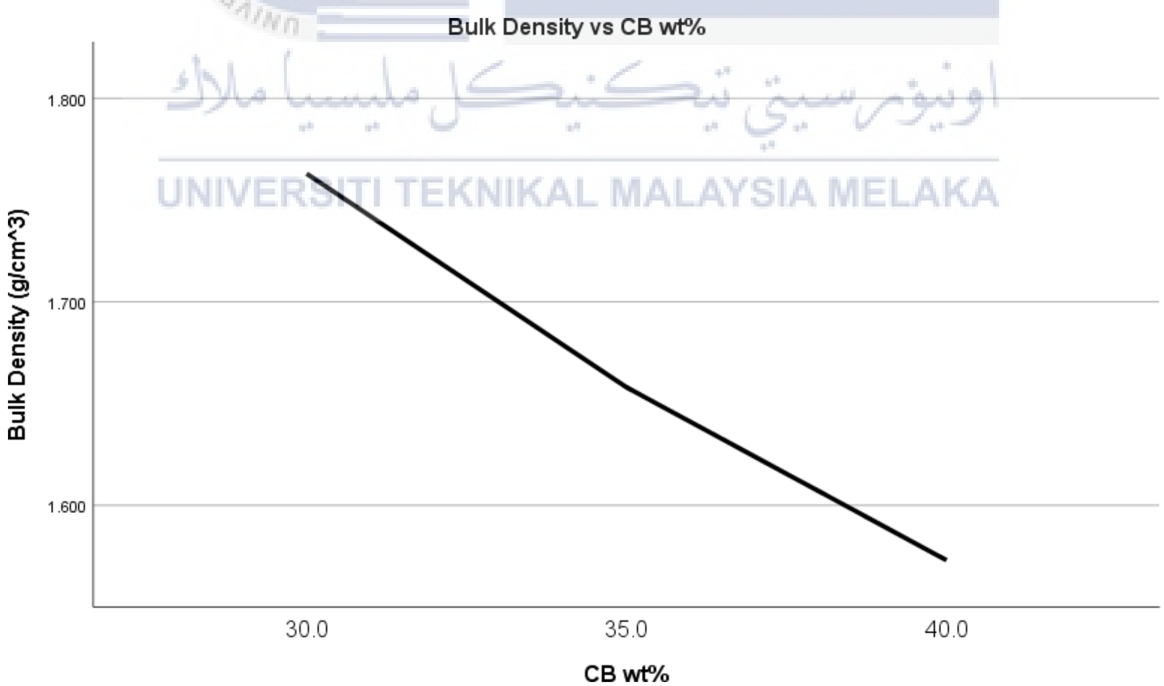

Figure 4.6 Graph of Bulk Density vs CB wt%

4.6 COORDINATE MEASUREMENT TEST

Several key dimensions labeled as No. 1 – 5 of the flow channel of Sample 1, 3 and 5 are measured and compared with dimensions of flow channel drawing as shown in Figure 4.7.

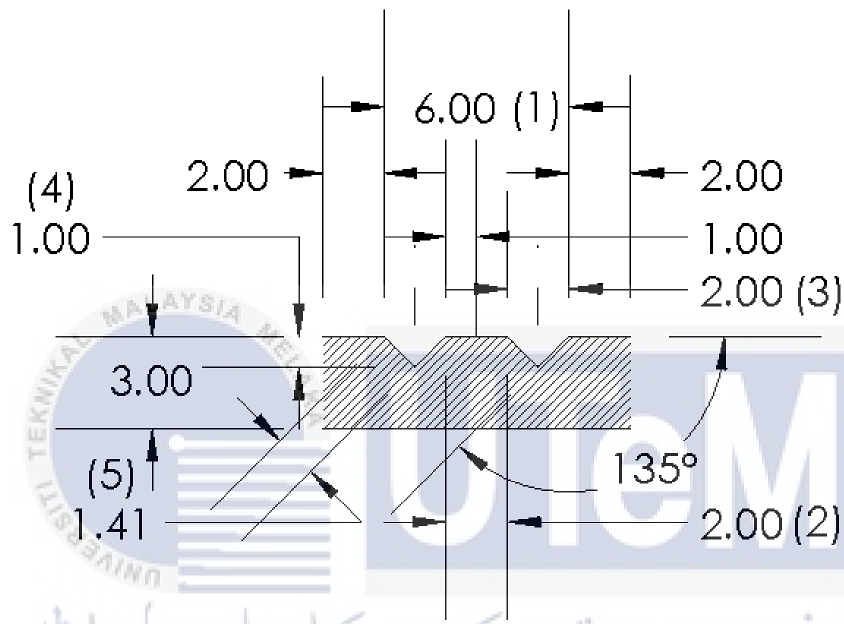

Figure 4.7 Dimensions No. 1 – 5 of Flow Channel Drawing

Table 4.7 Percentage Difference for Dimension No. 1 – 5 for Sample 1, 3 and 5 and Drawing

Dimension No.	Drawing Dimension (mm)	Sample 1 Dimension (mm)	Sample 3 Dimension (mm)	Sample 5 Dimension (mm)	Average Dimension for Samples 1, 3 & 5 (mm)	Percentage Difference between Drawing and Average Dimension (%)
1	6	6.083	6.105	6.096	6.0947	1.578
2	2	2.035	1.985	2.052	2.0240	1.2
3	2	1.988	2.011	1.943	1.9807	0.965
4	1	1.008	0.989	1.028	1.0083	0.83
5	1.41	1.416	1.409	1.414	1.4130	0.213

From Table 4.7, the percentage difference between drawing and average dimension of the 3 flow channels fabricated ranged from the minimum 0.213% for dimension No. 5 to maximum of 1.578% for dimension No. 1.

CHAPTER 5

DISCUSSION

5.1 ELECTRICAL CONDUCTIVITY ANALYSIS

From the graph in Figure 4.3 shown, the electrical conductivity increased with the amount of carbon black weight percentage. This is expected due to higher concentration of conductive fillers will filled up more spaces in the material and formed more extensive conductive path. However, the graph also showed that the increment in conductivity is getting smaller when carbon black weight percentage increased to 40 wt%. This suggests the percolation threshold for the composite's electrical conductivity is not far from 40 wt% carbon black. Any increment in carbon black weight percentage above that level brings no significant increase in electrical conductivity.

5.2 FLEXURAL STRENGTH ANALYSIS

From the graph in Figure 4.4 shown, the flexural strength increased with the amount of carbon black weight percentage until 35 wt%, further increased to 40 wt% of carbon black resulted in drop of flexural strength. This indicated that the initial introduce of carbon black into the polymer matrix will strengthen the interfacial bonding between the particles. However, the concentration of carbon black when exceed percolation threshold, the content of epoxy resin will decrease to a level that resulted in weaker interconnectivity between the particles of resin and fillers. This resulted in fillers cannot be fully wetted and welly aggregated among the composite, causing weaker interaction between the particles.

5.3 HARDNESS ANALYSIS

From the graph in Figure 4.5 shown, the shore hardness increased with the amount of carbon black weight percentage. This is probably due to higher hardness of carbon black particles compared to epoxy and graphite particles causing it more resist to indentation. Besides, incorporation of carbon black particles filled the void and decreased the inter-particle distance, causing the composite more resistant to indentation.

5.4 BULK DENSITY ANALYSIS

From the graph in Figure 4.6 shown, the bulk density almost decreased linearly with the amount of carbon black weight percentage. Carbon black has lower density than graphite and epoxy. When content of carbon black increased, the overall density of composite decreased. The decreasing density also due to the porosity developed in the composites during process of fabrication. The 3 composition samples tested have density between 1.5 – 1.8 g/cm^3, which are below the value stipulated by US DOE which is 1.90 g/cm^3. This implied that they are suitable materials for fabrication of bipolar plate due to the light weight advantage.

5.5 COORDINATE MEASUREMENT ANALYSIS

From Table 4.7, the dimension accuracy of the 5 measured dimensions range from 0.213 to 1.578 %. The 2 largest deviations from the drawing are the dimension No. 1 and 2. This is due to the samples when taken out from the mold experienced minor damages along the flow channel ribs such as broken at some areas and scratches. These conditions had translated into some differences in dimensions with the drawing. Nevertheless, the differences in dimensions measured are bearable, hence the V-shaped Serpentine flow field is suitable to be fabricated by hot press method.

5.6 DIFFICULTIES IN EXPERIMENT

5.6.1 Samples Fabricated Adhered to Mold

In first attempt, during the process to take out the fabricated samples from the mold, especially the mold with flow channel, it is discovered that the sample stick to the mold surface tightly. If separated by brute force, the sample will break around the flow channel as shown in Figure 5.1. This scenario is considered popular problem faced by using hot compression method since the mold is pressed by high pressure causing the material stick to the mold surface. The solution to solve the problem is to make sure the mold surface is clean entirely, followed by applying lubricating oil on the mold surface. This help to reduce the friction between material and mold interface, ease the process to take out the sample from the mold.

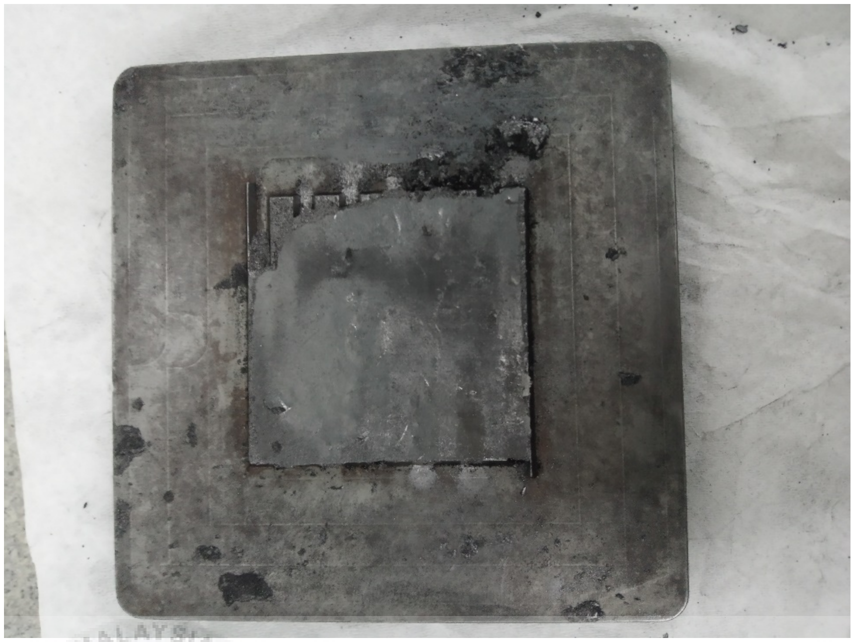

Figure 5.1 Unsuccessful First Attempt in Fabrication of Flow Channel

5.6.2 Wear and Tear of Molds

The molds have numerous scratches and slightly bended at corners. This poses difficulty when putting the upper mold onto the material due to the tight dimension and some degree of differences in geometry. The solution to the problem is to use a soft hammer to knock the upper mold slightly into the cavity. The rest of the part can be pressed by the hydraulic press of the hot press machine to fully allow the upper mold pressed onto the materials.

CHAPTER 6

CONCLUSION AND RECOMMENDATION

6.1 CONCLUSION

In this study, the effect of carbon black on the properties of graphite/epoxy composite and the parameter dimension of flow channel is examined. Total 6 samples of flow and cooling channels had been fabricated with hot compression molding. Out of the three different composition of carbon black used, testing results showed that 35 wt% is the best composition since in this proportion, the sample tested approaches threshold in electrical conductivity testing, showed highest flexural strength, has intermediate hardness and low in bulk density. Adding of carbon black filler at this weight percentage level does increase electrical conductivity, hardness and flexural strength of the composite, while decreasing its bulk density. V-shape Serpentine flow field is fabricated on the surface of the flow channel and the dimension of sample flow channel measured showed that the difference between them and drawing are not obvious which is within 1.6%. This result showed that hot compressing method is a promising method for fabrication of V-shaped Serpentine flow field flow channel.

6.2 RECOMMENDATION

The result of this experiment is useful to identify the best composition for best performance when a bipolar plate is made up of graphite/carbon black/epoxy materials. In future research, the range of composition tested shall be increased to a wider range with narrowing gap between each consecutive weight percentage. This approach helps to identify more accurate critical weight percentage where turning point occur for a characteristics tested. Different types of fillers such as carbon nanotube and different matrix component such as polypropylene are worth to be explored to compared their performance with carbon black filler and epoxy matrix.

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APPENDIX A

ELECTRICAL CONDUCTIVITY

Area, A = 100 mm², Gage Length, L = 2.5 cm

SAMPLE 2 (Gr/CB/EP: 45/30/25)

Orientation	Position	I (mA)	V (mV)
Front	**1**	1	0.055
	2	1	0.048
	3	1	0.052
	4	1	0.051
	5	1	0.036
	6	1	0.051
	7	1	0.049
	8	1	0.042
	9	1	0.049
Back	**1**	1	0.039
	2	1	0.041
	3	1	0.046
	4	1	0.038
	5	1	0.040
	6	1	0.049
	7	1	0.051
	8	1	0.043
	9	1	0.044
		$V_{\bar{x}}$	0.0458

$R_{\bar{x}}$ = 45.8 m Ω

ρ = 183.2 μ Ω m

SAMPLE 4 (Gr/CB/EP: 40/35/25)

Orientation	Position	I (mA)	V (mV)
Front	**1**	1	0.033
	2	1	0.040
	3	1	0.039
	4	1	0.042
	5	1	0.046
	6	1	0.038
	7	1	0.039
	8	1	0.043
	9	1	0.042
Back	**1**	1	0.037
	2	1	0.038
	3	1	0.042
	4	1	0.040
	5	1	0.031
	6	1	0.036
	7	1	0.035
	8	1	0.043
	9	1	0.036
		$V_{\bar{x}}$	0.0389

$R_{\bar{x}}$ = 38.9 m Ω

ρ = 155.6 μ Ω m

SAMPLE 6 (Gr/CB/EP: 35/40/25)

Orientation	Position	I (mA)	V (mV)
Front	1	1	0.043
	2	1	0.037
	3	1	0.030
	4	1	0.034
	5	1	0.031
	6	1	0.042
	7	1	0.038
	8	1	0.035
	9	1	0.030
Back	1	1	0.035
	2	1	0.039
	3	1	0.038
	4	1	0.031
	5	1	0.035
	6	1	0.038
	7	1	0.036
	8	1	0.039
	9	1	0.032
		$V_{\bar{x}}$	0.0357

$R_{\bar{x}}$ = 35.7 m Ω

ρ = 142.8 μ Ω m