

**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
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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 any 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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SELAMAT

Date :

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 grafit dan juga karbon hitam dicampur dengan epoksi sebagai pengikat untuk menghasilkan komposit plat dwikutub yang konduktif. Komposisi pengisi dan pengikat 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 menggunakan 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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LIST OF ABBREVIATIONS

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°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.

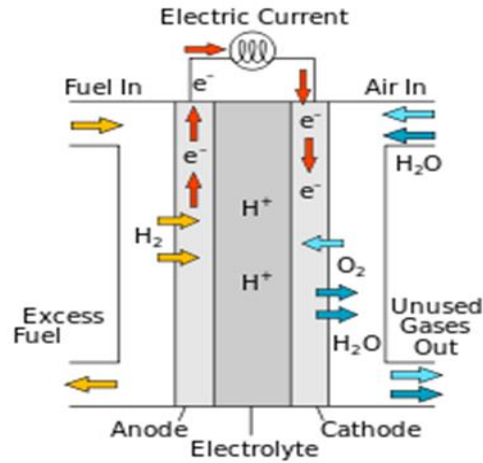


Figure 2.1: Basic fuel cell schematic diagram (Source: Wikipedia)

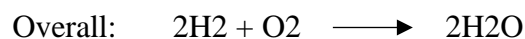
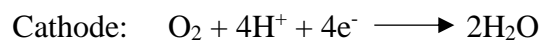
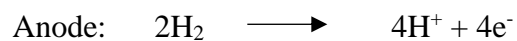
Table 2.1: Characteristics of different type of fuel cells (Source: Ismail, (2011))

		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:



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.

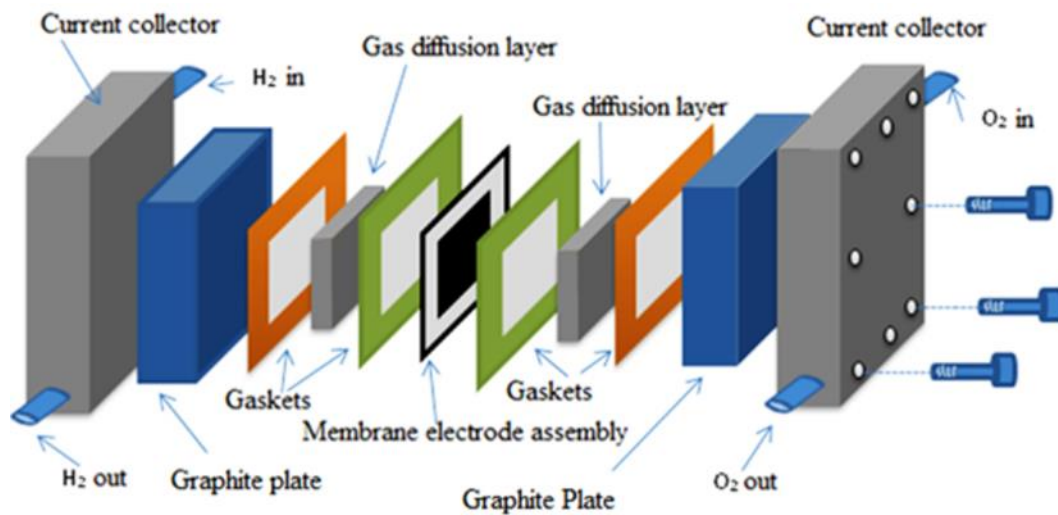


Figure 2.2: Exploded view of PEMFC main component

(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

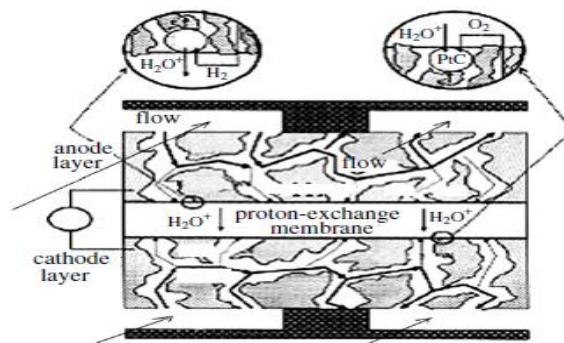


Figure 2.3: Structure of a membrane electrode assembly (MEA). The arrangement of principal units between gas flow along BP channels and flows in MEA

(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	$\Omega-cm^2$	0.02	0.01
Flexural strength	MPa	>25	>25