EFFECT OF FILLERS ON BIPOLAR PLATE POLYPROPYLENE COMPOSITES FOR PEM FUEL CELL

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ผลของสารตัวเติมที่มีต่อแผ่นนำไฟฟ้าสองขั้วพอลิพรอพิลีนคอมโพสิตสำหรับเซลล์เชื้อเพลิงพีอีเอ็ม

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ABSTRACT

This research was prepared bipolar plate from polymer composite using for polymer electrolyte membrane fuel cells (PEMFCs). It was studied the effect of conductive polymer composite with various ratio of PP and conducting fillers (Acetylene black (AB), Carbon black N330 (CBn), Carbon black ENSACO 250 (CBe), Graphite RGN-C (GT), Carbon nanotubes (CNTs)) and then they were compared with acid treated fillers. They were blended by two-roll mill and were ground with grinding machine. After the various ratios of composites were compressed, then were analyzed by density, water absorption, mechanical properties, scanning electron microscopy, interfacial contact resistance and electrical conductivity. It was found that polymer composites with conductive fillers and treated with acid treatment, the electrical resistance was decreased. The polymer composites given the highest electrical conductivity was PP:ATAB:ATCBe:CNT(N7000) in a ratio of 50:23:23:4. The polymer composites were dominated the electrical conductivity of in-plane technique about 50.51 S/cm, electrical conductivity of 47.12 S/cm in through-plane, density of 1.38 g/cm^3, water absorption of 0.108 % , hardness of 79.5 HRD and flexural strength of 25.35 MPa, respectively.

Keywords: polymer composite, bipolar plate, polymer electrolyte membrane fuel cells, polypropylene, conductive fillers

หัวข้อวิทยานิพนธ์	ผลของสารตัวเติมที่มีต่อแผ่นนำไฟฟ้าสองขั้วพอลิพรอพิลีน คอมโพสิตสำหรับเซลล์เชื้อเพลิงพีอีเอ็ม
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บทคัดย่อ

งานวิจัยนี้ได้ศึกษาการเตรียมแผ่นนำกระแสไฟฟ้าแบบสองขั้ว จากพอลิเมอร์คอมพอสิต เพื่อใช้ในเซลล์เชื้อเพลิงชนิดเยื่อแลกเปลี่ยนโปรตอน โดยทำการเตรียมแผ่นพอลิเมอร์คอมพอสิตจากหลายอัตราส่วนของพอลิพรอพิลีน และ สารตัวเติมนำไฟฟ้า (อะเซทีลีนแบล็ค คาร์บอนแบล็คชนิดN330 คาร์บอนแบล็คชนิดENCASO250 แกรไฟท์ชนิดRGN-C คาร์บอนนาโนทิวป์) จากนั้นนำมาเปรียบเทียบกับสารตัวเติมนำไฟฟ้าที่ถูกปรับสภาพโดยกรด ทำการผสมแต่ละอัตราส่วนโดยใช้เครื่องผสมสองลูกกลิ้ง และทำการบด หลังจากนั้นนำเม็ดคอมพอสิตในแต่ละอัตราส่วนไปทำการกดอัด โดยใช้เครื่องกดอัดและทำการวิเคราะห์สารตัวอย่างเพื่อหาความหนาแน่น ความสามารถในการดูดซับน้ำ สมบัติเชิงกล สัณฐานวิทยา ค่าความต้านทานเชิงสัมผัส และค่าการนำไฟฟ้า พอลิเมอร์คอมพอสิตที่มีค่าการนำไฟฟ้ามากที่สุดคือ พอลิพรอพิลีนคอมโพสิตที่ประกอบด้วย พอลิพรอพิลีน ร้อยละ 50 อะซิทิลีนแบล็คที่ผ่านการปรับสภาพโดยกรดแล้วร้อยละ 23 คาร์บอนแบล็คชนิดENSACO250ที่ปรับสภาพแล้วร้อยละ 23 และ ท่อคาร์บอนนาโนชนิด N7000ร้อยละ 4 โดยมีค่าการนำไฟฟ้าในแนวแกน50.51 ซีเมนส์ต่อเซนติเมตร ค่าการนำไฟฟ้าในแนวทะลุผ่าน 47.12 ซีเมนส์ต่อเซนติเมตร ความหนาแน่น 1.38 กรัมต่อลูกบาศก์เซนติเมตร ความสามารถในการดูดซับน้ำร้อยละ 0.108 ค่าความแข็ง 79.5 HRD และค่าความแข็งแรงดัด 25.35 เมกกะพาสคาล ตามลำดับ

คำสำคัญ : พอลิเมอร์คอมพอสิต แผ่นนำไฟฟ้าสองขั้ว พอลิพรอพิลีน สารตัวเติมนำไฟฟ้า

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Miss Anotai Saowamol

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Chapter 1

Introduction

1.1 Motivation

Energy sources are very important for human life. Human need differences types of energy in our life to perform a different task. Human gets energy from various natural resources that are non-renewable such as petroleum, coal and natural gas. The energy consumption by source was shown in Figure 1.1

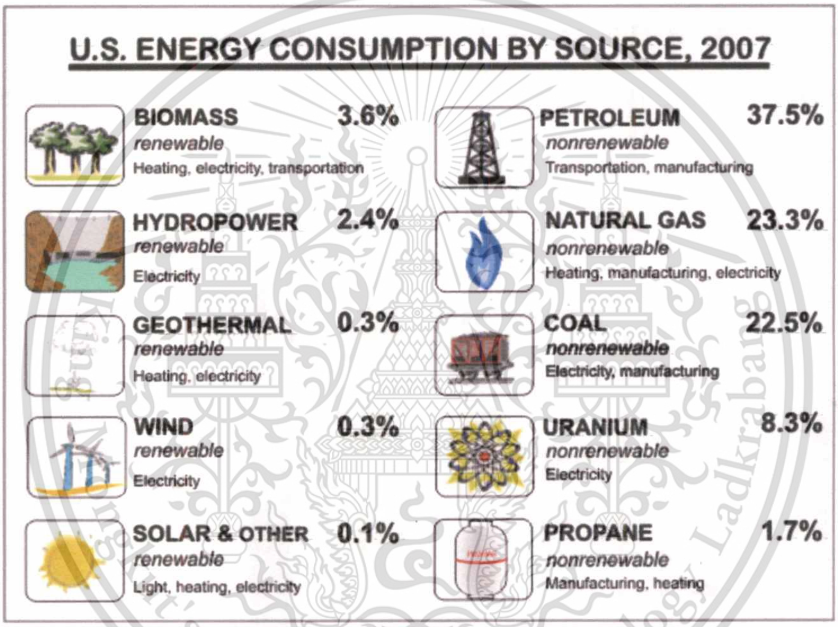


Figure 1.1 U.S. energy consumption by source [1]

The need for energy sources is getting necessary, hence oil and gas price are steadily rising, and there has been a steady movement towards developing higher fuel efficiency and more alternative fuel vehicles. There are many reasons to looking towards to alternative energy sources. Many countries were effort to reduce pollutants and greenhouse gases. Alternative energy source show significant promise for reduction the amount of toxics which comes from by-products of energy use and preserve many of the natural resource that we currently use as sources of energy. The interest alternative energy is fuel cell.

to cell and prevents leakages of gases and coolant.

Furthermore, bipolar plate accounts 80% weight and 60% cost of PEM fuel cell stack. Therefore, it is very probably the most expensive component in an electrochemical cell stack because of the high costs of fabrication plates and machining the fluid-flow channels. Hence, cost reduction of bipolar plate is necessary. In order to achieve the design criteria, function and cost targets of the bipolar plate, researchers tend to focus on the material selection of bipolar plate. According to these benefits of fuel cell, this research aims to study the preparation of polymer composite bipolar plate having good carbon polymer. Bipolar plates have many advantages, such as high corrosion resistance, low contact resistance and low fabrication cost, due to the easy fabrication method. Composite bipolar plates have also higher electrical conductivity which is improved by increasing the filler content in the composite. However, these fillers are difficult to apply to bipolar plates due to of their poor mechanical properties. Therefore, this research was also study the parameter effecting to electrical conductivity properties of bipolar plate prepared by carbon composite mixing with polypropylene. In addition, the present study is focused on polymer composites containing conductive fillers such as carbon black N330, carbon black ENSACO 250, acetylene black, graphite RGN-C, carbon nanotubes(N7000) and carbon nanotubes(Timesnano) having unique characteristics such as high electrical conductivity, large specific surface area and strong adsorptive ability that will provide good efficiency of bipolar plate. [4]

1.2 Objectives

1. To prepare bipolar plate from polymer composites between Polypropylene(PP) matrix and various ratio of conducting fillers (Carbon black(CB), Acetylene black(AB), Graphite RGN-C(GT), Carbon black ENSACO 250(CBh), Carbon nanotubes(CNTs), acid treated carbon black N330(ATCB), acid treated acetylene black(ATAB), acid treated graphite RGN-C (ATGT) and acid treated carbon black ENSACO 250 (ATCBe)) by using two-roll mill and compression molding.

2. To study properties of bipolar plate that prepared from polymer composites between Polypropylene (PP) matrix and various ratio of conducting fillers (Carbon black N330 (CBn), Acetylene black (AB), Graphite (GT), Carbon black ENSACO 250(CBe), Carbon

nanotubes(CNTs), acid-treated carbon black N330 (ATCB), acid treated acetylene black(ATAB), acid treated graphite RGN-C (ATGT) and acid treated carbon black ENSACO 250(ATCBe))

1.3 Scope of this study

1. To prepare bipolar plate from polymer composites between Polypropylene (PP) matrix and various ratios of conducting fillers

2. To determine physical properties such as water absorption and density of polymer composites.

3. To determine mechanical properties such as flexural strength and hardness of polymer composites.

4. To determine electrical conductivity followed by ASTM C611 test procedure (Standard test method for electrical resistivity of polymer composites).

5. To determine morphology of polymer composites by Scanning Electron Microscope (SEM).

1.4 Expected results

1. To have good performance of electrical conductivity of bipolar plate.

2. To possibly investigate bipolar plate for industrial proton exchange membrane fuel cells in the future.

Chapter 2

Theory and Literature Reviews

2.1 Technology of fuel cell

Consumption of energy (or useful energy, exergy, to be thermodynamically correct) has become a daily necessity in the modern world, dramatically increasing along with improvements to the quality of life, the industrialization of developing nations, and the increase of world population. At present, the majority of energy required worldwide is met by combustion of fossil fuels. These resources have become an essential and integral part of modern life, increasingly relied upon since the Industrial Revolution. It has long been recognized that the excessive energy consumption has a significant adverse impact on the environment, resulting in increased health risks to all life forms and the threat of global climate change. The diminishing fossil fuel reserve is also intensifying international tensions, transforming national security, and contributing to high inflation. Fuel cell technology has the potential to meet the extraordinary energy needs of our modern civilization and lessen the undesirable effects of energy consumption [5].

Fuel cells are being developed at an increasingly fast pace with many academic researchers entering the field. Fuel cells are environmentally friendly devices for energy conversion and power generation, and are one of the most promising candidates as a zero-emission power source. Hence, they are often regarded as one of the advanced energy technology of the future. In reality, fuel cells are one of the oldest energy conversion devices known to humankind, although their development and deployment for practical applications lag far behind other competitive technologies, mainly heat engines employing combustion of fossil fuels have become an essential and integral part of modern civilization, being increasingly relied upon since the Industrial Revolution [6].

A fuel cell is an electrochemical cell that converts chemical energy from a fuel into electric energy. We may define a fuel cell as an energy conversion device for power generation that similar to batteries and heat engines. In principle, a fuel cell operates like a battery. But, a battery is really an energy storage device. So, the maximum amount of useful energy from a battery is determined by the amount of chemical reactants stored within the battery itself. Therefore, the lifetime of a battery is quite limited. In comparison, a fuel cell can produce electric energy output for as long as the fuel and oxidant are supplied. Because the reactants are stored externally, the amount of useful energy derivable from a fuel cell is inexhaustible as long as the reactants are available. On the other hand, a heat engine – driven generator can also convert the chemical energy of reactants into electric energy but, it use a multistep energy – conversion process involving several intermediate energy forms and conversion processes in several different devices. Although, both fuel cell and heat engines are energy conversion devices with reactants stored externally, fuel cells operate isothermally at given temperature and electric energy is generated directly in one step through electrochemical reactions of the reactants without any intermediate forms of energy involved. Therefore, it might be expected that fuel cells will have higher electrical energy efficiency than heat engines [7].

Fuel cell is composed of a fuel electrode (anode), an oxidant electrode (cathode), and electrolyte fuel cell as shown in figure 2.1

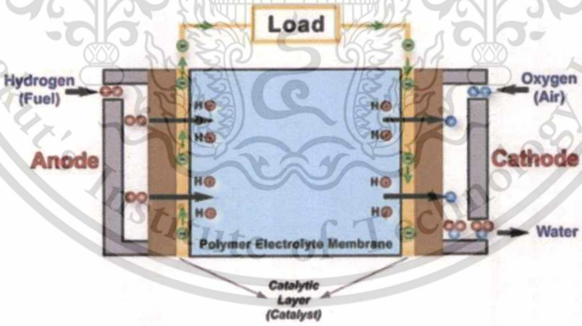


Figure 2.1 The operating principle of the polymer electrolyte membrane fuel cell. [8]

Molecular hydrogen is delivered from a gas flow stream to one of the electrode (anode) then hydrogen reacts electrochemically in the anode as follow equation. (2.1)

Anode half – cell reaction: $H_2 \longrightarrow 2\,H^+ + 2e^-$ (2.1)

Hydrogen (fuel) is oxidized at the anode/electrolyte interface into hydrogen ion or proton H^+ and gives up electron e^-. The protons migrate through the electrolyte (acid) and electrons are forced to transfer through an external circuit. Then, the electrons and protons both arrive at cathode. At the cathode, the protons and electrons react with the oxygen supplied from and external gas flow steam to forming water as equation

Cathode half – cell reaction: $\frac{1}{2}O_2 + 2e^- + 2H^+ \longrightarrow H_2O$ (2.2)

Thus, oxygen is reduced into water at cathode by combining with H^+ and e^-. Now the electric current and mass transfer forms a complete circuit. As a result, the overall cell reaction can be obtain by summing the above two half – cell reaction to overall cell reaction as equation

Overall cell reaction: $H_2 + O_2 \longrightarrow 2H_2O$ (2.3)

The fuel cell offers several advantages more than other types of power generation device, such as, high efficiency, high power density, low operating temperature and mild pressure conditions, simple construction and low pollutant. There are several types of fuel cells for example, Proton Exchange Membrane Fuel Cell (PEMFCs), Phosphoric Acid Fuel Cells (PAFCs), Alkaline Fuel Cells (AFCs), Molten Carbonate Fuel Cells (MCFCs), and Solid Oxide Fuel Cell (SOFC). The PEMFCs operates at low temperature and allowing faster start – ups and immediate response to charges in power demand. It is the most promising system compare to the other types of fuel cells for the automotive industry [9].

2.2 Types of fuel cell [9]

2.2.1 **Proton Exchange Membrane Fuel Cells (PEMFC)**

The proton exchange membrane fuel cell (PEMFC) is one of the most elegant types of fuel cells in terms of its design and mode of operation. It consists of solid polymeric proton conducting membrane (the electrolyte), which is sandwiched between two platinum catalyzed porous gas diffusion electrodes in single cell. The proton exchange membrane fuel cell was the first type of fuel cell to find an application as an auxiliary power source in NASA's Gemini Space Vehicles in the early 1960s. Because of the problems of relatively low power densities ($<100mW/cm^2$) and short life times (< 2 weeks) caused by the low performing proton conducting membrane, the PEMFC power source was replaced by an alkaline fuel cell (AFC) system for NASA's Apollo and space Shuttle flights from the mid 1960s until the present time. PEMFCs have attained the highest performance levels and longest lifetimes of all types of fuel cells. The PEMFC system is also unique in that it has the technical capabilities of covering a wide range of power levels. Since the late 1980s, the momentum has been rising exponentially to develop PEMFCs and a multitude of advancements have been made in PEMFC R&D, resulting in several thousand publications. PEM fuel cells use a solid polymer membrane (a thin plastic film) as the electrolyte. This polymer is permeable to protons when it is saturated with water, but it does not conduct electrons as shown in Figure 2.2

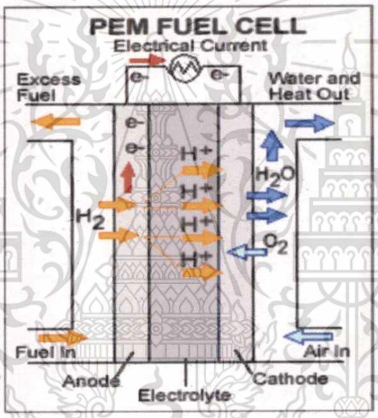


Figure 2.2 Proton Exchange Membrane Fuel Cells (PEMFC) [9]

The fuel for the PEMFC is hydrogen and the charge carrier is the hydrogen ion (proton). At the anode, the hydrogen molecule is split into hydrogen ions (protons) and electrons. The hydrogen ions permeate across the electrolyte to the cathode while the electrons flow through an external circuit and produce electric power. Oxygen, usually in the form of air, is supplied to the cathode and combines with the electrons and the hydrogen ions to produce water. The reactions at the electrodes are as follows:

$$2H_2 \longrightarrow 4H^{\oplus} + 4e^{-} \qquad (2.4)$$

$$O_2 + 4H^{\oplus} + 4e^{-} \longrightarrow 2H_2O \qquad (2.5)$$

Compared to other types of fuel cells, PEMFCs generate more power for a given volume or weight of fuel cell. This high-power density characteristic makes them compact and lightweight. In addition, the operating temperature is less than 100ºC, which allows rapid start-up. These traits and the ability to rapidly change power output are some of the characteristics that make the PEMFC the top candidate for automotive power applications.

Other advantages result from the electrolyte being a solid material, compared to a liquid. The sealing of the anode and cathode gases is simpler with a solid electrolyte, and therefore, less expensive to manufacture. The solid electrolyte is also more immune to difficulties with orientation and has fewer problems with corrosion, compared to many of the other electrolytes, thus leading to a longer cell and stack life.

2.2.2 Alkaline Fuel Cells (AFC)

The first alkaline electrolyte used in fuel cell was molten KOH in the 19th century. The fuel cell was solid carbon and oxygen was the cathodic reactant. This was followed by the work of francis Bacon,whose goal was develop a 5 kW AFC system using pure hydrogen and oxygen as the reactants, This development and demonstration was completed over a 20 years. The electrolyte in the Bacon cell was molten and the cell operating temperature and pressure were 200 ºC and 50 atm., respectively. In an alkaline fuel cell, water is produced on the anode side, according to the equation:

$$H_2 + 2OH^- \rightarrow 2H_2O + 2e^-_o \qquad (2.6)$$

The cathodic reaction is:

$$1/2O_2 + H_2O + 4e^-_o \rightarrow 2OH^- \qquad (2.7)$$

Alkaline fuel cells (AFC) operate around 120°C to150°C using an aqueous solution of potassium hydroxide (KOH) as the electrolyte. Desirable attributes of the alkaline fuel cell include its high performance compared to other fuel cells and its flexibility to use a wide range of electrocatalysts. However, AFCs are intolerant to CO_2 which reacts with the KOH and effectively degrades the cell performance. Even the smallest amount of CO_2 in the oxidant would have to be scrubbed when considering the alkaline cell. This constraint requires that pure hydrogen and pure oxygen, not air, be used. [9]

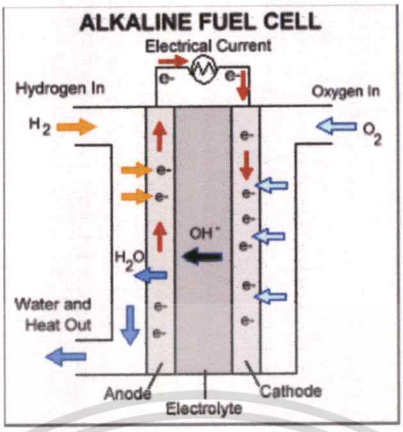


Figure 2.3 Alkaline fuel cells (AFCs) [9]

2.2.3 Phosphoric Acid Fuel Cells (PAFC)

The PAFC system was the first fuel cell technology to be commercialized for civilian applications. In FAFC, the acid serves as the ionic electrolyte as well as the solvent. The ionization reaction of the acid is represented by the equation:

$$H_3PO_4 \longrightarrow H^+ + H_2PO_4 \tag{2.8}$$

This reaction is followed by the salvation of the acid according to the equation:

$$H^+ + H_3PO_4 \longrightarrow H_4PO_4^+ \tag{2.9}$$

The charge carrier in this type of fuel cell is the hydrogen ion (H^+, proton). This is similar to the PEFC where the hydrogen introduced at the anode is split into its protons and electrons. The protons migrate through the electrolyte and combine with the oxygen, usually from air, at the cathode to form water. The electrons are routed through an external circuit where they can perform useful work. This set of reactions in the fuel cell produces electricity and by-product heat.

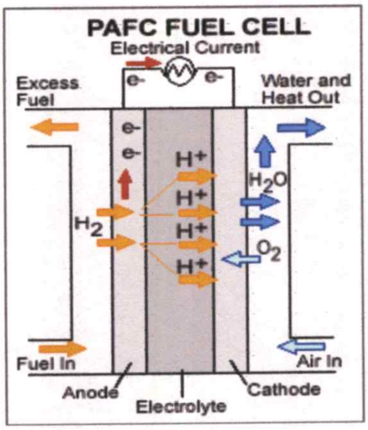


Figure 2.4 Phosphoric acid fuel cells [9]

2.2.4 Molten Carbonate Fuel Cells (MCFC)

MCFC power plants are capable of direct utilization of natural gas and coal derived gases for electric and thermal power generation at high efficiencies. In an MCFC, the cathodic reaction consumes oxygen and carbon dioxide to produce carbonate ions, which are transported to the anode through the electrolyte. At the anode, hydrogen combines with the carbonate ion to produce water and carbon dioxide. Carbon dioxide is then shuttled back to the cathode for the cathodic reaction. A schematic of the single cell in an MCFC is shown in Figure 2.4. This electrolyte has a reasonable ionic conductivity only at temperatures above 600oC. The current operating temperature is 650oC. At temperatures above 700oC, there is some loss of electrolyte by vaporization. One adventage of the high operating temperature of MCFCs is that activation overpotentials for the fuel cell reactions are considerably lower than those for the low and intermediate temperature fuel cells. This type of fuel cell was considered the second – generation fuel cell for power generation (the first being the PAFC). But MCFC technology has faced many challenges, particularly because of the relatively high corrosion rates of component materials in the electrolyte at the high operating temperatures.

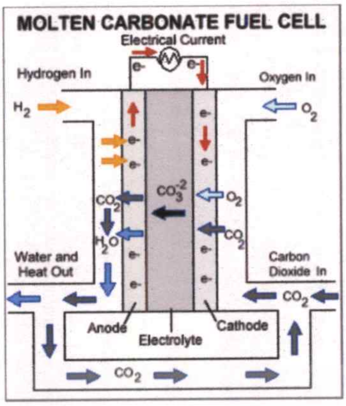


Figure 2.5 Molten Carbonate Fuel Cells [9]

2.2.5 Solid Oxide Fuel Cell (SOFC)

The Solid Oxide Fuel Cell (SOFC) is currently the highest-temperature fuel cell in development and can be operated over a wide temperature range from 600ºC–1000ºC allowing a number of fuels to be used. To operate at such high temperatures, the electrolyte is a thin, solid ceramic material (solid oxide) that is conductive to oxygen ions (O^{2-}). The SOFC has been in development since the late 1950s and has two configurations that are being investigate-planar (flat panel) and tubular.

As a solid electrolyte, it is impervious to gas cross-over from one electrode to another when liquid electrolytes usually consist of the electrolyte contained in some porous supporting structure. The charge carrier in the SOFC is the oxygen ion (O^{2-}). At the cathode, the oxygen molecules from the air are split into oxygen ions with the addition of four electrons. The oxygen ions are conducted through the electrolyte and combine with hydrogen at the anode, releasing four electrons. The electrons travel an external circuit providing electric power and producing by-product heat.

$$\text{Anode Reaction} \quad : \quad 2H_2 + 2O^{2-} \rightarrow 2H_2O + 4e^- \tag{2.10}$$

$$\text{Cathode Reaction :} \quad O_2 + 4e- \rightarrow 2O^{2-} \tag{2.11}$$

$$\text{Overall Cell Reaction :} 2H_2 + O_2 \rightarrow 2H_2O \tag{2.12}$$

The very high operating temperature of the SOFC has both advantages and disadvantages. The high temperature enables them to tolerate relatively impure fuels, such as those obtained from the gasification of coal or gasses from industrial process and other sources. However, the high temperatures require more expensive materials of construction.

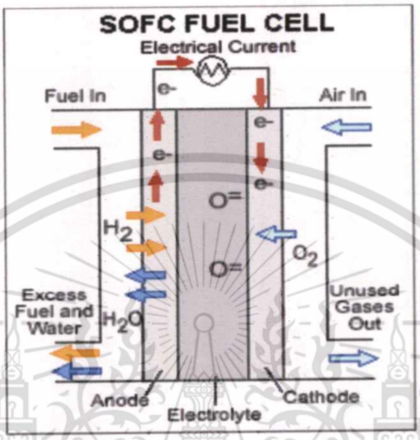


Figure2.6 Solid Oxide Fuel Cell (SOFC) [9]

2.3 Bipolar plate [10]

Polymer electrolyte membrane fuel cells (PEMFCs) are able to efficiently generate high power densities, making the technology potentially attractive for certain mobile and portable applications. Bipolar plate being the most important components in PEMFCs stacks because its account for the bulk of the stack (in both weight and volume) and are used as support to distribute fuel (hydrogen) and air and carry generated electrons. Accordingly, bipolar plates require the following high mechanical strength, electrical conductivity and corrosion resistance, low volume and weight, and low fabrication cost. Bipolar plate works by current anode/cathode plates produced by adding conductive elements to polymers. For this project, study to preparation of conductive polymer composites and efforts using carbon black (CB), carbon brush (CBh), acetylene black (AB), carbon nanotubes (CNTs), Graphite (GT), acid treated carbon black (ATCB) and acid treated carbon brush (ATCBh) filled conducting polymers.

Composite materials are formed by combining two or more materials that have quite different properties. The different materials work together to give the composite unique properties, but within the composite you can easily tell the different materials apart they do not dissolve or blend into each other.

2.4 Composites [11]

Most composites are made up of just two materials. One material (the matrix or binder) surrounds and binds together a cluster of fibers or fragments of a much stronger material (the reinforcement). In the case of mud bricks, the two roles are taken by the mud and the straw; in concrete, by the cement and the aggregate; in a piece of wood, by the cellulose and the lignin. In fiberglass, the reinforcement is provided by fine threads or fibers of glass, often woven into a sort of cloth, and the matrix is a plastic.

Polymer composites have 2 phase

1. Matrix material

Matrix is the monolithic material into which the reinforcement is embedded, and is completely continuous. This means that there is a path through the matrix to any point in the material, unlike two materials sandwiched together.

2. Reinforcement material

Reinforcement material is embedded into the matrix. The reinforcement does not always serve a purely structural task (reinforcing the compound), but is also used to change physical properties such as wear resistance, friction coefficient, or thermal conductivity. The reinforcement can be either continuous, or discontinuous. Discontinuous MMCs can be isotropic, and can be worked with standard metalworking techniques, such as extrusion, forging or rolling. In addition, they may be machined using conventional techniques, but commonly would need the use of polycrystaline diamond tooling (PCD).

Continuous reinforcement uses monofilament wires or fibers such as carbon fiber or silico carbide. Because the fibers are embedded into the matrix in a certain direction, the result is an anisotropic structure in which the alignment of the material affects its strength. One of the first MMCs used boron filament as reinforcement. Discontinuous reinforcement uses "whiskers", short fibers, or particles. The most common reinforcing materials in this category are alumina and silicon carbide.

2.5 Materials for Bipolar plate

2.5.1 Polypropylene [12]

We can divide plastic that use to make bipolar plate into thermoplastic and thermosetting plastics (thermosets). Thermoplastic is a plastic that melts to a liquid when heated and freezes to a brittle, very glassy state when cooled sufficiently. Most thermoplastics are high-molecular-weight polymers whose chains associate through weak Van der Waals forces (polyethylene); stronger dipole-dipole interactions and hydrogen bonding (nylon); or even stacking of aromatic rings (polystyrene). Thermoplastic polymers are difference from thermosetting polymers (Bakelite; vulcanized rubber) as they can, unlike thermosetting polymers, be remelted and remolded. Many thermoplastic materials are addition polymers; e.g., vinyl chain-growth polymers such as polyethylene and polypropylene.

Thermosetting plastics (thermosets) are polymer materials that irreversibly cure to a stronger form. The cure may be done through heat (generally above 200°C), through a chemical reaction (two-part epoxy, for example), or irradiation such as electron beam processing.

Thermoset materials are usually liquid or malleable prior to curing and designed to be molded into their final form, or used as adhesives. Others are solids like that of the molding compound used in semiconductors, and Integrated Circuits (IC's). The curing process transforms the resin into a plastic or rubber by a cross-linking process. Energy and/or catalysts are added that cause the molecular chains to react at chemically active sites (unsaturated or epoxy sites, for example), linking into a rigid, 3-D structure. The cross-linking process forms a molecule with a larger molecular weight, resulting in a material with a higher melting point. During the reaction, when the molecular weight has increased to a point so that the melting point is higher than the surrounding ambient temperature, the material forms into a solid material.

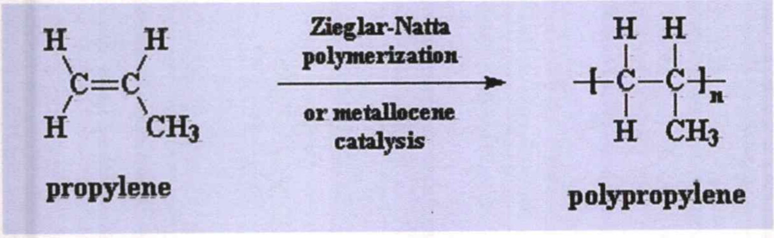
Uncontrolled reheating of the material results in reaching the decomposition temperature before the melting point is obtained. Therefore, a thermoset material cannot be melted and re-shaped after it is cured. This implies that thermosets cannot be recycled, except as filler material.

Thermoset materials are generally stronger than thermoplastic materials due to this 3-D network of bonds, and are also better suited to high-temperature applications up to the decomposition temperature of the material.

The difference between thermoplastics and thermosetting plastics is that thermoplastics become soft, remoldable and weldable when heat is added. Thermosetting plastics however can't be welded or remolded when heated, simply burning instead. On the other hand, once a thermosetting is cured it tends to be stronger than a thermoplastic. For this research used polypropylene as a matrix of polymer composites.

Polypropylene is one of those rather versatile polymers out there. It serves double duty, both as a plastic and as a fiber. As a plastic it's used to make things like dishwasher-safe food containers. It can do this because it doesn't melt below 160°C, or 320°F. Polyethylene, a more common plastic, will anneal at around 100°C, which means that polyethylene dishes will warp in the dishwasher. As a fiber, polypropylene is used to make indoor-outdoor carpeting, the kind that you always find around swimming pools and miniature golf courses. It works well for outdoor carpet because it is easy to make colored polypropylene, and because polypropylene doesn't absorb water, like nylon does.

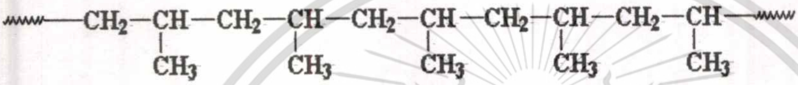
Structurally, it's a vinyl polymer, and is similar to polyethylene, only that on every other carbon atom in the backbone chain has a methyl group attached to it. Polypropylene can be made from the monomer propylene by Ziegler-Natta polymerization and by metallocene catalysis polymerization.



(2.13)

$$H_2C{=}CH(CH_3) \xrightarrow[\text{or metallocene catalysis}]{\text{Zieglar-Natta polymerization}} \left[\underset{H}{\overset{H}{C}}-\underset{CH_3}{\overset{H}{C}}\right]_n$$

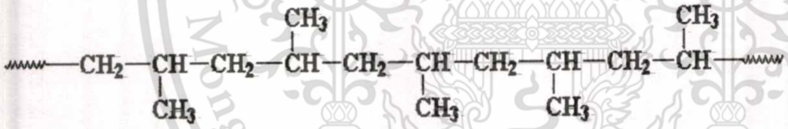
Research is being conducted on using metallocene catalysis polymerization to synthesize polypropylene. Metallocene catalysis polymerization can do some pretty amazing things for polypropylene. Polypropylene can be made with different tacticities. Most polypropylene we use is isotactic. This means that all the methyl groups are on the same side of the chain, like this



$$\sim CH_2-\underset{CH_3}{CH}-CH_2-\underset{CH_3}{CH}-CH_2-\underset{CH_3}{CH}-CH_2-\underset{CH_3}{CH}-CH_2-\underset{CH_3}{CH}\sim$$

isotactic polypropylene (2.14)

But sometimes we use atactic polypropylene. Atactic means that the methyl groups are placed randomly on both sides of the chain like this:



$$\sim CH_2-\underset{CH_3}{CH}-CH_2-\overset{CH_3}{CH}-CH_2-\underset{CH_3}{CH}-CH_2-\underset{CH_3}{CH}-CH_2-\overset{CH_3}{CH}\sim$$

atactic polypropylene (2.15)

However, by using special metallocene catalysts, it's believed that we can make polymers that contain blocks of isotactic polypropylene and blocks of atactic polypropylene in the same polymer chain, as is shown in the figure below:

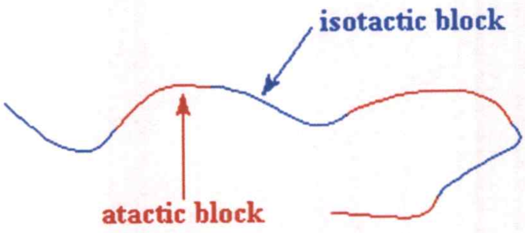


Figure 2.7 Blocks of isotactic polypropylene and block of atactic polypropylene in the same polymer chain [12]

This polymer is rubbery, and makes a good elastomer. This is because the isotactic blocks will form crystals by themselves. But because the isotactic blocks are joined to the atactic blocks, the little hard clumps of crystalline isotactic polypropylene are tied together by soft rubbery ethers of atactic polypropylene.

To be honest, atactic polypropylene would be rubbery without help from the isotactic blocks, but it wouldn't be very strong. The hard isotactic blocks hold the rubbery isotactic material together, to give the material more strength. Most kinds of rubber have to be crosslinked to give them strength, but not polypropylene elastomers.

Elastomeric polypropylene, as this copolymer is called, is a kind of thermoplastic elastomer. However, until the research is completed, this type of polypropylene will not be commercially available.

The polypropylene that you can buy off the shelf at the store today has about 50 - 60% crystallinity, but this is too much for it to behave as an elastomer (Figure 2.8).

Figure 2.8 Polypropylene [13]

Typical Polypropylene Applications are used for Automotive Applications, Household Goods, Film, Appliances, Containers, Packaging, Industrial Applications, Electrical/Electronic Applications, and Closures

Advantages of Polypropylene are Homopolymer, Processability, Food Contact Acceptable, Impact Resistance, Stiffness, Copolymer, Flow, Impact Resistance and Chemically Coupled but disadvantages of Polypropylene are degraded by UV, Flammable (retarded grades available), Attacked by chlorinated solvents and aromatics, Difficult to bond, several metals accelerate oxidative degrading and Low temperature impact strength is poor.

2.5.2 Carbon black [13]

Carbon black is a material produced by the incomplete combustion of heavy petroleum products such as tar, coal tar, ethylene cracking tar, and a small amount from vegetable oil. Carbon black is a form of amorphous carbon that has a high surface area to volume ratio, and as such it is one of the first nanomaterials to find common use, although its surface area to volume ratio is low compared to activated carbon. It is similar to soot but with a much higher surface area to volume ratio. Carbon black is used as a pigment and reinforcement in rubber and plastic products.

1. Production

Two carbon black manufacturing processes (furnace black and thermal black) produce nearly all of the world's carbon blacks, with the furnace black process being the most common. The furnace black process uses heavy aromatic oils as feedstock. The production furnace uses a closed reactor to atomize the feedstock oil under carefully controlled conditions (primarily temperature and pressure). The primary feedstock is introduced into a hot gas stream (achieved by burning a secondary feedstock, e.g., natural gas or oil) where it vaporizes and then pyrolyzes in the vapor phase to form microscopic carbon particles. In most furnace reactors, the reaction rate is controlled by steam or water sprays. The carbon black produced is conveyed through the reactor, cooled, and collected in bag filters in a continuous process. Residual gas, or tail gas, from a furnace reactor includes a variety of gases such as carbon monoxide and hydrogen. Most furnace black plants use a portion of this residual gas to produce heat, steam, or electric power.

The thermal black process uses natural gas, consisting primarily of methane or heavy aromatic oils, as feedstock material. The process uses a pair of furnaces that alternate approximately every five minutes between preheating and carbon black production. The natural gas is injected into the hot refractory lined furnace, and, in the absence of air, the heat from the refractory material decomposes the natural gas into carbon black and hydrogen. The aerosol material stream is quenched with water sprays and filtered in a bag house. The exiting carbon black may be further processed to remove impurities, pelletized, screened, and then packaged for shipment. The hydrogen off-gas is burned in air to preheat the second furnace.

2. Physical & Chemical Properties

Carbon black is not soot or black carbon, which are the two most common, generic terms applied to various unwanted carbonaceous by-products resulting from the incomplete combustion of carbon-containing materials, such as oil, fuel oils or gasoline, coal, paper, rubber, plastics and waste material. Soot and black carbon also contain large quantities of dichloromethane and toluene extractable materials, and can exhibit an ash content of 50% or more.

Carbon black is chemically and physically distinct from soot and black carbon, with most types containing greater than 97% elemental carbon arranged as aciniform (grape-like cluster) particulate. On the contrary, typically less than 60% of the total particle mass of soot or black carbon is composed of carbon, depending on the source and characteristics of the particles (shape, size, and heterogeneity). In the case of commercial carbon blacks, organic contaminants such as polycyclic aromatic hydrocarbons (PAHs) can only be extracted under very rigorous laboratory analytical procedures (soxhlet extraction using organic solvents and high temperatures). These extracts, though they may be similar to those derived from soot, are unique, however, because carbon black extracts exist only in extremely small quantities. Water and body fluids are ineffective in removing PAHs from the surface of carbon black and, therefore, they are not considered to be biologically available. Two other commercial carbonaceous products often confused with carbon black are activated carbon and bone black. Each is produced by processes different from commercial carbon black and possesses unique physical and chemical properties. The structure of carbon black was shown in Figure 2.9

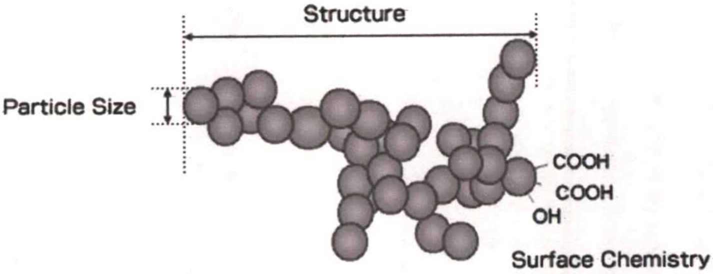


Figure 2.9 Structure of carbon black [13]

3. Carbon black uses

Traditionally, carbon black has been used as a reinforcing agent in tires. Today, because of its unique properties, the uses of carbon black have expanded to include pigmentation, ultraviolet (UV) stabilization and conductive agents in a variety of everyday and specialty high performance products, including:

Tires: and Industrial Rubber Products: Carbon black is added to rubber as both a filler and as a strengthening or reinforcing agent. For various types of tires, it is used in innerliners, carcasses, sidewalls and treads utilizing different types based on specific performance requirements. Carbon black is also used in many molded and extruded industrial rubber products, such as belts, hoses, gaskets, diaphragms, vibration isolation devices, bushings, air springs, chassis bumpers, and multiple types of pads, boots, wiper blades, fascia, conveyor wheels, and grommets.

Plastics: Carbon blacks are now widely used for conductive packaging, films, fibers, moldings, pipes and semi-conductive cable compounds in products such as refuse sacks, industrial bags, photographic containers, agriculture mulch film, stretch wrap, and thermoplastic molding applications for automotive, electrical/electronics, household appliances and blow-molded containers.

Electrostatic Discharge (ESD) Compounds: Carbon blacks are carefully designed to transform electrical characteristics from insulating to conductive in products such as electronics packaging, safety applications, and automotive parts.

High Performance Coatings: Carbon blacks provide pigmentation, conductivity, and UV protection for a number of coating applications including automotive (primer basecoats and clearcoats), marine, aerospace, decorative, wood, and industrial coatings. Toners and Printing Inks: Carbon blacks enhance formulations and deliver broad flexibility in meeting specific color requirements.

Type of carbon black that use in this thesis

1.) Carbon black N330

Carbon black N330 is the most widely used type high wear-resisting of carbon black, the wear-resisting performance than super wear-resistant series black is a bit poor, but better than black. The slot method, Used to tire surface, pulp, sidewall and all kinds of rubber industry products. This product is a kind of carbon, can give good strong stretched rubber, resisting avulsion intensity, abrasion and elastic. Using this product by use of the rolling loss (loss) in behind only N300 series black N351 greater than any other species are small and scattered in the rubber, and the pressure of a performance is good, is suitable for various kinds of synthetic rubber and natural rubber.

2.) Carbon black ENSACO 250 conductive carbon blacks are carbon blacks with a high to very high void volume allowing the retention of a carbon network at low to very low filler content. The void volume can originate from the interstices between the carbon black particle due to their complex arrangement and from the porosity.

2.5.3 Acetylene black

Acetylene black is a form of carbon black formed by the decomposition (but not by the burning) of acetylene. It is characterized by its high degree of crystalline orientation compared with other sources of carbon black. Acetylene black is used as follows a component of depolarisation mixtures in dry cell batteries, as a pigment with an antistatic effect in plastics, an electrically conducting filler in coatings covering conductors and in field boundary layers in high-voltage cables, an electrically conducting filler with antistatic properties in parts made of technical rubber, a carbon component in hard metal carbides for metallurgical applications, a carbon component in electrocarbons and a pigment in electrically conducting paints. [14] Structure of acetylene black was shown in Figure 2.10

Figure 2.10 Structure of acetylene black (AB) [14]

2.5.4 Carbon nanotubes

Carbon nanotubes (CNTs) are allotropes of carbon with a cylindrical nanostructure. Nanotubes have been constructed with length-to-diameter ratio of up to 132,000,000:1, significantly larger than any other material. These cylindrical carbon molecules have novel properties, making them potentially useful in many applications in nanotechnology, electronics, optics, and other fields of materials science, as well as potential uses in architectural fields. They may also have applications in the construction of body armor. They exhibit extraordinary strength and unique electrical properties, and are efficient thermal conductors. Nanotubes are members of the fullerene structural family, which also includes the spherical buckyballs. The ends of a nanotube may be capped with a hemisphere of the buckyball structure. Nanotubes are categorized as single-walled nanotubes (SWNTs) and multi-walled nanotubes (MWNTs). [15] The structure of carbon nanotube was shown in Figure 2.11

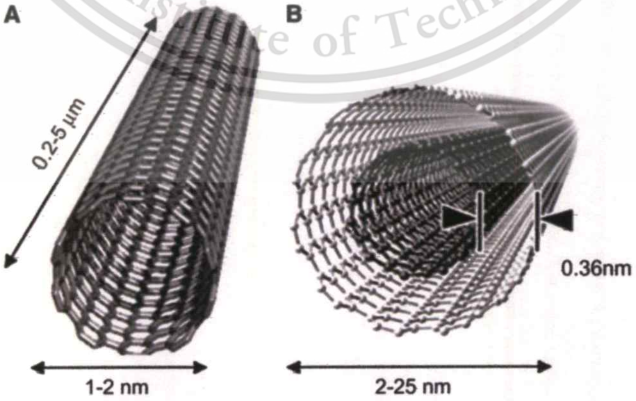


Figure 2.11 Structure of carbon nanotubes a.) single wall carbon nanotubes b.) multiwall carbon nanotubes [16]

Their carbon nanotubes name is derived from their size, since the diameter of a nanotube is on the order of a few nanometers (approximately 1/50,000th of the width of a human hair), while they can be up to 18 centimeters in length (as of 2010). However, a small diameter of carbon nanotubes can influence strongly electrical properties [16]. The figure of CNTs has shown in Figure 2.12.

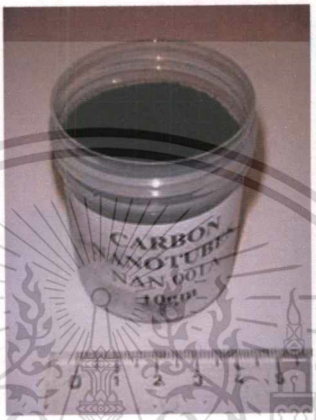


Figure2.12 Carbon nanotubes powder [17].

In this research, we focus on multiwall carbon nanotubes(timesnano) and multiwall carbonnanotubes(Nanocyl®N7000).

1.) Multiwall carbon nanotubes(timesnano) When cylinders are assembled one inside another, they are called a multi-wall carbon nanotube (MWNT). The dimensions of CNTs range from a few nanometers to dozens of nanometers in diameter and from the micron scale up to the mm scale in length. This geometry imparts a very high aspect ratio, which is critical to enhance most properties of polymer composites. MWNT produced by chemical vapour deposition are the most common type of carbon nanotubes. [18] Properties of carbon nanotubes (timesnano) was shown in table 3.6

2.) Multiwall carbonnanotubes(Nanocyl®N7000) is thin multi-wall carbon nanotubes, These carbon nanotubes are unique and prized world-wide because their small size and high aspect ratio (>150) lets them form a network of conductivity at a very low concentration. It is produced in multi-tons, via the Chemical Vapor Deposition (CVD) process. Nanocyl is installing a new reactor with a capacity of 400 tpa for producing its NC 7000 carbon nanotubes. Properties of carbon nanotubes(Nanocyl®N7000) was shown in table 3.7

2.5.5 Graphite [19]

Graphite is a polymorph of the element carbon. Diamond is another polymorph. The two share the same chemistry, carbon, but have very different structures and very different properties.

- Diamond is the hardest mineral known to man, Graphite is one of the softest.
- Diamond is an excellent electrical insulator, Graphite is a good conductor of electricity.
- Diamond is the ultimate abrasive, Graphite is a very good lubricant.
- Diamond is usually transparent, Graphite is opaque.
- Diamond crystallizes in the Isometric system and graphite crystallizes in the hexagonal system.

Somewhat of a surprise is that at surface temperatures and pressures, Graphite is the stable form of carbon. In fact, all diamonds at or near the surface of the Earth are currently undergoing a transformation into Graphite. This reaction, fortunately, is extremely slow. All of the differences between graphite and diamond are the result of the difference in their respective structures. Graphite has a sheet like structure where the atoms all lie in a plane and are only weakly bonded to the graphite sheets above and below. Diamond has a framework structure where the carbon atoms are bonded to other carbon atoms in three dimensions as opposed to two in graphite. The carbon-carbon bonds in both minerals are actually quite strong, but it is the application of those bonds that make the difference. It may seem strange that one of the softest minerals (and a very slippery lubricant) is the high-strength component in composites used to build automobiles, aircraft, and of course golf club shafts. It is the weakly bonded sheets that slide by each other to yield the slipperiness or softness. Yet when those sheets are rolled up into fibers, and those fibers twisted into threads, the true strength of the bonds becomes apparent. The threads are molded into shape, and held in place by a binder (such as an epoxy resin). The resulting composites have some of the highest strength-to-weight ratios of any materials (excluding, of course, diamond crystals and carbon nanotubes). Graphite can only be confused with the mineral molybdenite which is metallic bluish silver in color. However, molybdenite is much denser and has a silver blue streak.

Most graphite is produced through the metamorphism of organic material in rocks. Even coal is occasionally metamorphosed into graphite. Some graphite is found in igneous rocks and also as nodules inside of iron meteorites. Structure of Graphite was shown in Figure 2.13

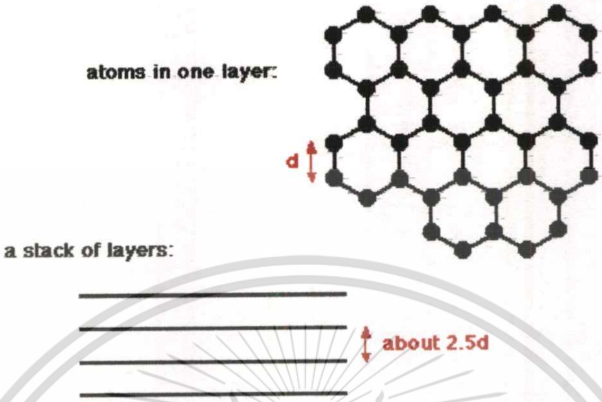


Figure 2.13 Structure of graphite [19]

Physical characteristic of graphite

- Color is black silver.
- Luster is metallic to dull.
- Transparency crystals are opaque
- Crystal System is hexagonal; 6/m 2/m 2/m
- Crystal Habits include massive lamellar veins and earthy masses. also as scaly granules in metamorphic rocks.
- Hardness is 1 - 2
- Specific Gravity is 2.2 (well below average)
- Cleavage is perfect in one direction.
- Fracture is flaky.
- Streak is black gray to brownish gray.
- Associated Minerals include quartz, calcite, micas, iron meteorites and tourmalines.
- Other Characteristics: thin flakes are flexible but inelastic, mineral can leave black marks on hands and paper, weakly conducts electricity.

- Notable Occurrences include New York and Texas, USA; Russia; Mexico; Greenland and India.
- Best Field Indicator is softness, luster, density and streak.

2.6 Composite fabrication

2.6.1 Two – roll mill [20]

A laboratory two-roll mill is used to hand-process plastics and the like. It looks like an old fashioned "wringer" from a washing machine mounted horizontally, but it isn't. The rolls counter-rotate, to be sure, but they do so at different rates to create a shearing action in the trough formed between them. In this case, one roll is turning faster than the other, because one drive sprocket has 16 teeth and the other has 22. Plastics are generally very poor at thermal conductivity, so this shearing action is very important to achieve a melt.

The rolls are heated internally with electrical heat rods, and the mill is designed to have a safe operating range up to 300 degrees F. No external heater, or pump, or special thermal fluid is required, and there is nothing to leak.

The rolls themselves are made from stainless steel. The thick walls give the rolls a great deal of thermal inertia, and facilitate maintaining the temperature accurately with the attached controller. Temperature is measured directly from the rolls with a radiant-energy sensor adjacent to blackened areas at the ends of the rolls.

Shearing plastics to melt them requires a lot of force. The rolls of our mill are driven at roughly one revolution every two seconds by a 2.2 Horsepower, three phase motor (220-240 volts/60 HZ) through a heavy-duty speed-reducing gearbox. No plastic has ever stalled this machine, or even slowed it perceptively. The weak link in this system is actually the drive chains, and you can see how massive they are (Figure 2.14).

Figure2.14 Operation of two-roll mill [Lab tech]

The rolls are generously sized at six inches in diameter, and have a nineteen inch working length. There is an additional two inches on the end of each roll that is blackened for temperature monitoring.

Our two-roll mill is reversible, which facilitates cleaning, and may be stopped almost instantly with either of two "knee" bars mounted on either side of the mill. It should be obvious from the pictures that this machine could be very dangerous in the wrong hands. The drive chains are exposed to enable them to be greased easily, and the mill rolls could easily pass a human hand through or worse — with terrible effect. For this reason, no sale will be made without a signed liability waiver. In other words, if you buy it and does something which gets you hurt with it, we are in no way responsible.

2.6.2 Compression molding

Compression molding is a method of molding in which the molding material. The mold is closed with a top force or plug member, pressure is applied to force the material into contact with all mold areas, while heat and pressure are maintained until the molding material has cured. The process employs thermosetting resins in a partially cured stage, either in the form of granules or preforms.[20] Compression molding is a high-volume, high-pressure method suitable for molding complex, high-strength fiberglass reinforcements. Advanced composite thermoplastics can also be compression molded with unidirectional tapes, woven fabrics, randomly oriented fiber mat or chopped strand. The advantage of compression molding is its ability to mold large, fairly intricate parts. Also, it is one of the lowest cost molding methods compared with other methods such as transfer molding and injection molding. Moreover, it wastes relatively little material, giving it an advantage when working with expensive compounds. However, compression molding often provides poor product consistency and difficulty in controlling flashing, and it is not suitable for some types of parts. Compression-molding is also suitable for ultra-large basic shape production in sizes beyond the capacity of extrusion techniques. [21]

Molding material, usually preheated, is placed into the open mold cavity. Once the mold is closed, heat and pressure are applied to help ensure that the mold is completely filled and as part of the curing process. [22] Schematic of compression molding has shown in Figure 2.15.

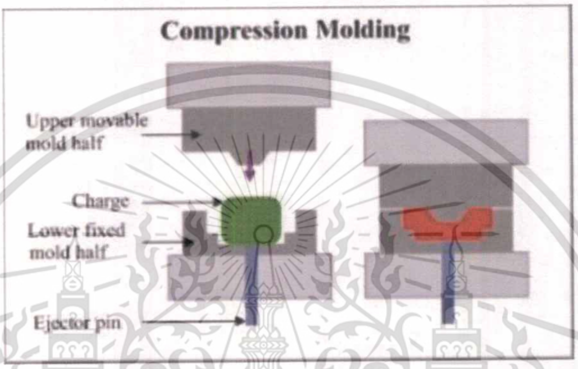


Figure 2.15 Schematic of compression molding [22]

Advantages of Compression Molding are lowest cost molds, little "throw away" material provides advantage on expensive compounds and often better for large parts but the disadvantages of Compression molding are Offers least product consistency, Difficult to control flash and not suited for some types of parts. [23]

2.7 Related literature

J. Auangdow [24] had prepared bipolar plate materials for proton exchange membrane fuel cell with polymer composites and conductive fillers composites. The study of the morphology, electrical conductivity, physical and mechanical properties of polymer composite prepared in this work showed that synergistic effect of acetylene black and carbon fiber in polymer composites provided higher electrical conductivity than that of the single filler in polymer composites. The polymer composites of acetylene black 20 wt% with carbon fiber 30 wt%, acetylene black 30 wt% with carbon fiber 20 wt% and acetylene black 40 wt% with carbon fiber 10 wt% possess the highest electrical conductivity of 3.3 S/cm. The cost evaluation of polymer composites showed that polymer composites of acetylene black 40 wt% with carbon fiber 10 wt% had the lowest cost. The conductivity of polymer composites can be improved by adding zinc sterate 5 phr and titanium dioxide 3 phr in polymer composites acetylene black 40 wt% with carbon fiber 10 wt%. It provides the best electrical conductivity of 5 S/cm. Zinc sterate and titanium dioxide increased the density and hardness, but decreased the water absorption and flexural strength of polymer composites.

S.R. Dhakate [25] had produced bipolar plate by compression molding technique using natural graphite, synthetic graphite, carbon fiber and carbon black as reinforcing constituents and phenolic resin as a binder matrix. A judicious combination and their respective proportions could produce a composite plate with bulk density 1.8–1.90 g/cm^3, electrical resistivity between 0.002 and 0.007 Ω·cm, shore hardness 65, flexural strength 45 MPa and flexural modulus 12 GPa. The characteristics and the performance of the composite plate developed by us are compared with the commercially available bipolar plates. A power density of 500 mW/cm^2 was achieved at 1400 mA/cm^2 current density when the above composite plate was used as a bipolar plate in a unit fuel cell

D. Radwan [26] had prepared the electrical properties of polypropylene/graphite (PP/G) composites as prospective replacements for the traditional graphite bipolar plate in proton-exchange membrane fuel cells. The composites have relatively low electrical conductivities, i.e., up to 28 S cm^{-1} at 90 wt.% G. Combination of G with carbon black (CB) is an effective way to develop higher conductivity composites. The conductivity reaches 35 S cm^{-1} by combination of 25 wt.% CB and 55 wt.% G to 20 wt.% PP. This is five times the value at 80 wt. % G and 20 wt.% PP (7 S cm^{-1}): Two methods are mainly adopted for the preparation of composites, namely, melt compounding and solution blending. Solution blending of PP with conductive fillers followed by moulding of the dried powder leads to higher conductivities compared with those of melt-compounded composites. The combination of conjugated conducting polymers such as polyaniline (PANi) with the PP, G, and CB is also investigated. It is found that composites containing PANi have lower conductivities than those of the neat composites. This decrease in conductivity is attributed to the poor thermal stability of PANi.

M. Borah [27] had prepared composite from Expanded graphite (EG), which is synthesized by chemical intercalation of natural graphite and rapid expansion at high temperature. The expanded graphite synthesized in this study has an expansion ratio between 75–100 cc/gm. The composite bipolar plate with varying weight percentage of EG gives different bulk density, electrical conductivity, mechanical properties and air tightness. The critical weight percentage of filler content is 50 to achieve the desired electrical conductivity and mechanical properties of bipolar plate as per U.S. DOE targets. The composite bipolar plate with 50 wt% of EG gives bulk density of 1.50 g/cm3, electrical conductivity >120 S/cm, bending strength 54 MPa, modulus 6 GPa and shore hardness 50. I–V characteristic of a cell assembly with EG-based composite plates are similar with the performance of a cell with commercial composite plates. These lightweight bipolar plates reduced the volume and weight of ultimate fuel cell stack and helped in improving the fuel cell performance.

J. Hee Lee [28] had prepared composite materials by using graphite with a small vol.% of carbon black (CB), multi-walled carbon nanotubes (MWNTs) or carbon fibres (CF) in an epoxy resin. The electrical conductivity and flexural properties of the composites are measured as a function of the carbon conductive filler content. The highest electrical conductivity is observed at a total conducting filler content of 75 vol. %. The addition of a small amount of hybrid conducting filler enhances the electrical conductivity up to certain threshold, viz. 5 vol.% of CB, 2 vol.% ofMWNTs, and 7 vol.% of CF. Above these thresholds, the electric conductivity decreases with increasing filler content, due to the lack of sufficient resin to bind the fillers tightly. The hybrid filler system has better properties than the single filler system. The experimental results indicate that there is an optimum composition range with respect to electrical conductivity and mechanical properties. A maximum value of the electrical conductivity of 254.7 S.cm^{-1} was observed in a composite composed of 73 vol. % of graphite, 2 vol. % of MWNTs and 25 vol. % of epoxy resin.

J. Sahari [29] had made of polypropylene/graphite (PP/G) composites as prospective replacements for the traditional graphite bipolar plate in proton-exchange membrane fuel cells. The composites have relatively low electrical conductivities, i.e., up to 28 S cm^{-1} at 90 wt.% G. Combination of G with carbon black (CB) is an effective way to develop higher conductivity composites. The conductivity reaches 35 S cm^{-1} by combination of 25 wt. % CB and 55 wt.% G to 20 wt.% PP. This is five times the value at 80 wt. % G and 20 wt. % PP (7 $S.cm^{-1}$). Two methods are mainly adopted for the preparation of composites, namely, melt compounding and solution blending. Solution blending of PP with conductive fillers followed by moulding of the dried powder leads to higher conductivities compared with those of melt-compounded composites. The combination of conjugated conducting polymers such as polyaniline (PANi) with the PP, G, and CB is also investigated. It is found that composites containing PANi have lower conductivities than those of the neat composites. This decrease in conductivity is attributed to the poor thermal stability of PANi.

S. Sharma [30] had improved the properties of graphite–polymer composite bipolar plate by nanostructuring. This involves the incorporation of different vol. % of multiwall carbonnanotubes (MWNTs) in graphite–polymer composite bipolar plate. It has been found that by inclusion of 1 vol.% of MWNTs in graphite composite plate, the electrical and thermal conductivity of nanocomposite increased by 100%. The thermal conductivity of nanocomposite plate increases from 1 W/m K to 13 W/m K in throughplane and in-plane from 25 W/m K to 50 W/m K at 1 vol. % of MWNTs. This significant enhancement is due to the orientation of MWNTs in all the directions of composite, positive synergistic effect of MWNTs and heat transfer along the axis directions. However, bending strength of nanocomposite increases by 25% and maximum augmentation is in case of 1 vol. % of MWNTs. The improvement in conductivity of nanocomposite plate is due to an increase in the electron transfer ability within the composite plate which influences the I–V performance of ultimate fuel cell. These observations confirm that the optimal content of MWNTs is 1 vol. %, in graphite–polymer composite.

H. Suherman [31] had focused on using a conductive polymer composite (CPC) as a potential replacement for the conventional graphite bipolar plate used in polymer electrolyte membrane fuel cells (PEMFC). Based on the requirements established by the US Department of Energy (DOE), the in-plane electrical conductivity and flexural strength are required to be greater than 100 S/cm and 25 MPa, respectively. The high filler loading is needed to satisfy the high in-plane electrical conductivity. However, the high filler loading reduces the flexural strength and manufacturability of the composite. In this study, the composites were prepared by compounding using an internal mixer followed by compression moulding. The combination of 10 vol% carbon black (CB) as the second filler with synthetic graphite/epoxy (SG/EP) resulted in the following composite properties: 150 S/cm (in-plane conductivity), 55 S/cm (through-plane conductivity), and 38.8 MPa (flexural strength). Used as the second filler, the CB, which had a small-sized diameter, formed conductive networks that filled the voids between the SG and polymer matrix. The in-plane electrical conductivity and flexural strength of the CB/SG/EP composites at the optimum composition exceeded the requirement for bipolar plate applications.

S. Saranee [32] had prepared of bipolar plate from carbon composite containing conductive polyaniline. To increase the electrical conductivity of composite plate, carbon such as RGN, BDH and carbon brush (CB) was used to study the electrical conductivity and mechanical properties. The result showed that CB and BDH gave similar electrical conductivity (14.8 S/cm) and higher than RGN. It was founded that the electrical conductivity could be improved by treated CB with acid. The investigation of added polyaniline and polyaniline nanofiber showed that polyaniline nanofiber increased the electrical conductivity but decreased mechanical properties. Carbon composite was prepared by adding treated CB during the synthesis of polyaniline nanofiber for homogeneous mixture. This method can be improving electrical conductivity but decrease mechanical properties. However, the research indicated that the mechanical properties could be improved by additives such as carbon fiber and carbon nanotubes. The result showed that the additives could be increased electrical conductivity and mechanical properties. From the cell performance test, the mixture of composite plate was composed of polyaniline nanofiber 7 %wt., carbon fiber 20 %wt. and treated CB 60 %wt. The carbon composite bipolar plate gave the current density of 219 mA/cm^2 at 0.6 V but it was lower than the commercial graphite bipolar plate (383 mA/cm^2).

H. Suherman [33] had studied; the electrical conductivity of a G/epoxy (EP) composite (single filler) is only 50 S/cm (in-plane conductivity) at 80 wt% G. However, flexural strength is greater than 25 MPa. We were using carbon nanotubes (CNTs) as the second filler at a concentration of 5 wt% in a CNTs/G/EP nanocomposite resulted in the in-plane and through-plane electrical conductivity and flexural strength being 180 S/cm, 75 S/cm, and 45 MPa, respectively. The density of the CNTs/G/EP nanocomposite is also less than that of G/EP composite, which demonstrates that a total weight reduction is achievable.

Chapter 3

Experimental Details

In this experiment, we prepared various ratio of Polypropylene(PP) – Carbon black N330 (CBn) /Acetylene black (AB) / Graphite RGN-C (GT) / Carbon black ENSACO 250 (CBe) / acid treated carbon black N330 (ATCBn) / acid treated acetylene black (ATAB) / acid treated graphite RGN-C (ATGT) / acid treated carbon black ENSACO 250 (ATCBe) / CNTs composites by two-roll mill. The blends were ground with grinding machine. Furthermore, the blends were molded by compression molding. Density, water absorption, conductivity, morphology and mechanical properties were also characterized to understand the nature of various ratios of blends.

3.1 Chemicals

1. Polypropylene (PP) grade: P701J SCG Performance Chemicals Company Limited

The properties of polypropylene are shown in Table 3.1

Table 3.1 Properties of Polypropylene (PP)

Properties	Unit
Melt flow rate (230°C / 2.16 kg),	12 g / 10 min
Density	0.910 g/cm^3
Tensile strength at yield	320 kg/cm^2
Elongation at break	650 %
Flexural modulus	15500 kg/cm^2
Notched izod impact strength at 23°C	3 kg.cm/cm
Heat deflection temperature, at 4.6 kg/cm^2	110 °C

2. Carbon black N-330: Thai Carbon Black Public Co., Ltd.

The properties of carbon black N-330 are shown in Table 3.2

Table 3.2 Properties of Carbon black N330

Component of Carbon black N330	**Carbon black N330**
Purity (%)	94.8
Ash content (%)	0.7
Volatile matter	2
Moisture (%)	2.5
BET surface area (m^2/g)	71-85
Particle size (micrometers)	0.026 – 0.03
Density (g/cm^3)	0.320

3. Acetylene black HICON BLACK AB 50 P: IRPC Public Company Limited

The Properties of acetylene black are shown in Table 3.3

Table 3.3 Acetylene black

Component of Acetylene black	**Acetylene black**
Purity (%)	98
Ash content (%)	0.20
Volatile matter	2
Moisture (%)	0.3
BET surface area (m^2/g)	71.18
Particle size (micrometers)	11.86± 0.57
Density (g/cm^3)	0.085

4. Graphite RGN-C: Sutee united carbon Public Company Limited

The Properties of graphite are shown in Table 3.4

Table 3.4 Properties of Graphite RGN-C

Component of Graphite RGN-C	**Graphite RGN-C**
Purity (%)	82.6
Ash content (%)	15.1
Volatile matter	2
Moisture (%)	0.3
BET surface area (m^2/g)	11.77
Particle size(micrometers)	6.54±0.12
Density (g/cm^3)	0.345

5. Carbon black ENSACO 250 Granular: Origin eason paint Co , Ltd.

The properties of carbon black ENSACO 250 are shown in Table 3.5

Table 3.5 Properties of Carbon black ENSACO 250

Component of Carbon black ENSACO 250	**Carbon black ENSACO 250**
Purity (%)	99.79
Ash content (%)	0.01
Volatile matter	0.1
Moisture (%)	0.1
BET surface area (m^2/g)	65.6
Particle size (micrometers)	0.04
Density (g/cm^3)	0.178
BET surface area after treated by acid (m^2/g)	98.85

6. Carbon nanotubes : Timesnano

The properties of carbon nanotubes timesnano are shown in Table 3.6

Table 3.6 Properties of Carbon nanotubes (Timesnano)

Component of Carbon nanotubes	**Component of Carbon nanotubes**
Outside diameter (nm)	30-50
Inside diameter (nm)	5-10
Length (micrometer)	10-20
Purity (%)	95
Conductivity (S/cm)	>100

7. Carbon nanotubes : Nanocyl®7000

The properties of carbon nanotubes Nanocly®7000 are shown in Table 3.7

Table 3.7 Properties of Carbon nanotubes : Nanocyl®7000

Component of Carbon nanotubes	**Component of Carbon nanotubes**
Outside diameter (nm)	9.5
Inside diameter (nm)	4
Length (micrometer)	10
Purity (%)	90
Conductivity (S/cm)	125

8. Concentrate hydrochloric acid : Carlo erba reagents

9. Concentrate Nitric acid : QREC (Asia)

3.2 Instruments

1. Two-roll mill: Labtech , Model LRM 110

2. Compression molding: Labtech, Model MGLP-20A1

3. Grinder: Bosco, Model BG 3035

4. Balance: DENVER Instrument Company, Model No.4102

5. Scanning Electron Microscope (SEM): JEOL Ltd., Model JSM 5800LV

6. Oven: WTC binder 50/60 Hz

7. Potentiostat: Autolab company, Model PGSTATO

8. Hardness test: IDENTEC Company, Model 4150 AK

9. Overhead stirrer: KIKA works company, Model RW20.n

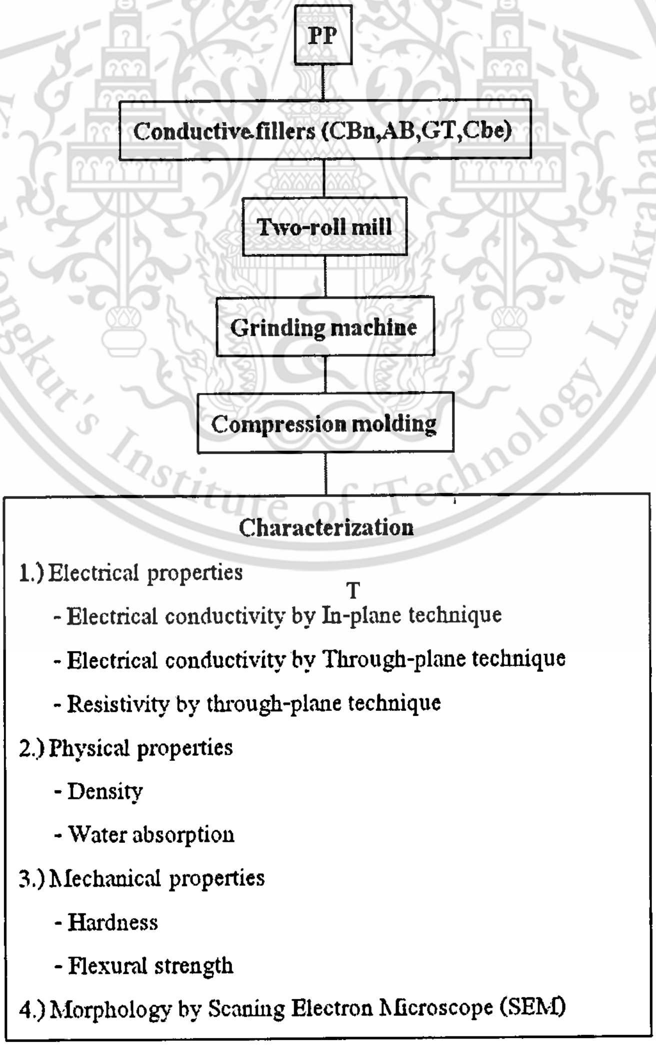
10. Four point probe: Jandael Engineering Limited, Model RM3-AR

11. Universal testing machine: LLOYD LR 10K PLUS

3.3 Flow chart of sample preparation

Sample preparation can be separated into three parts. First part was prepared polypropylene composite that contain of PP matrix and conductive fillers (CBn,AB,GT,CBe) in ratio of 50:50 and 50:25:25 as shown in Table 3.8-3.9. Second part was prepared polypropylene composite that contain of PP matrix and acid-treated conductive fillers (ATCBn,ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25 (Table 3.10-3.11). Third part was prepared polypropylene composite that contain of the optimum ratio of polypropylene composite from part 1 and 2 with CNTs(Timesnano) and CNT(N7000)(Table 3.12)

Part 1 Preparation of polypropylene composite that contain of polypropylene matrix and conductive fillers (CBn,AB,GT,CBe) in ratio of 50:50 and 50:25:25(Table 3.8-3.9).



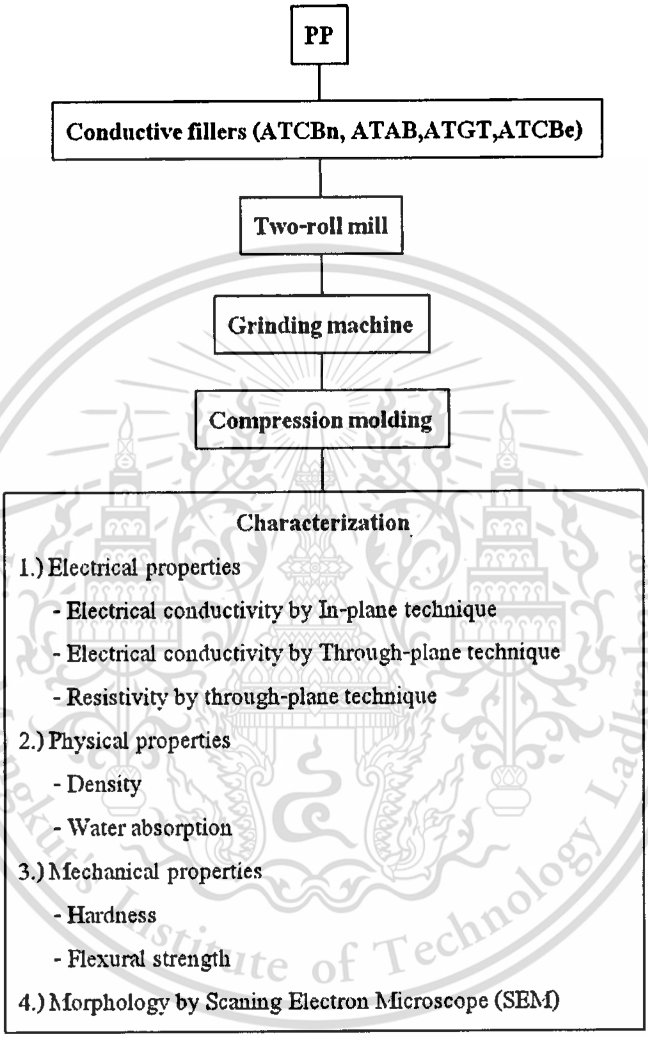
1.) Electrical properties
- Electrical conductivity by In-plane technique
- Electrical conductivity by Through-plane technique
- Resistivity by through-plane technique

2.) Physical properties
- Density
- Water absorption

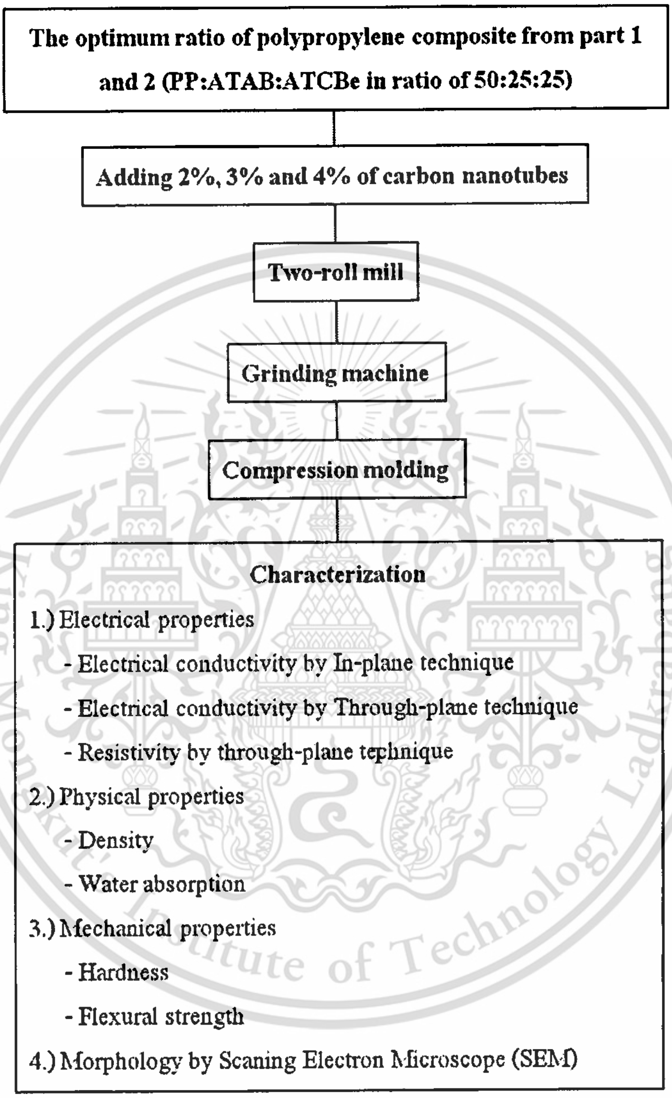
3.) Mechanical properties
- Hardness
- Flexural strength

4.) Morphology by Scaning Electron Microscope (SEM)

Part 2 Preparation of polypropylene composite that contain of Polypropylene matrix and acid-treated conductive fillers (ATCBn, ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25(Table 3.10 -3.11)



Part 3 Preparation of polypropylene composite that contain of the optimum ratio of polypropylene composite from part 1 and 2 with CNTs (Timesnano) and CNT (N7000) (Table 3.12)



3.4 Sample preparation

Sample preparation can be separated into three parts.

Part 1 Preparation of polypropylene composite that contain of polypropylene matrix and conductive fillers (CBn,AB,GT,CBe) in ratio of 50:50 and 50:25:25(Table 3.8-3.9).

The various ratios of Polypropylene (PP) matrix and conductive fillers were shown in Table 3.8 – 3.9, respectively.

Table 3.8 Preparation of Polypropylene matrix and conductive fillers (CBn,AB,GT,CBe) in ratio of 50:50

Polymer matrix	Conductive filler
50% by weight	50% by weight
PP	AB
PP	CBn
PP	GT
PP	CBe

Table 3.9 Preparation of Polypropylene matrix and conductive fillers (CBn,AB,GT,CBe) in ratio of 50:25:25

Polymer matrix	Conductive filler	Conductive filler
50% by weight	25% by weight	25% by weight
PP	AB	CBn
PP	AB	GT
PP	AB	CBe
PP	CBn	GT
PP	CBn	CBe
PP	GT	CBe

*Note: PP is Polypropylene, AB is Acetylene black, CBe is Carbon black ENSACO 250, AB is Carbon black N330, GT is Graphite RGN-C

Part 2 Preparation of polypropylene composite that contain of Polypropylene matrix and acid-treated conductive fillers (ATCBn, ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25 (Table 3.10 – 3.11)

Carbon black N330 (CBn), acetylene black (AB), Carbon black ENSACO 250 (CBe) and Graphite RGN-C (GT) were dried at 100 °C in a vacuum oven for 3 h to remove moisture. It was mixed with saturated acid consisting of sulfuric acid and concentrated nitric acid in a volume ratio of 3:1 for 10 h. Nitric acid serves as an oxidizer and sulfuric acid as an intercalant. The mixture was stirred from time to time to get the uniform intercalation of each flake. The mixture was carefully washed and filtrated with water until the pH level of solution reached 7 (neutral). After washing, acid-treated flakes were dried at 100 °C in a vacuum oven for 5 h. Keep the acid treated carbon for further experiment.

The various ratios of polypropylene (PP) matrix and acid treated conducting fillers were prepared as shown in Table 3.10 – 3.11, respectively

Table 3.10 Preparation of Polypropylene matrixes and acid treated conductive fillers (ATCBn, ATAB, ATCBe, ATGT) in ratio of 50:50

Polymer matrix	Acid treated conductive filler
50% by weight	50% by weight
PP	ATAB
PP	ATCBn
PP	ATGT
PP	ATCBe

Table 3.11 Preparation of Polypropylene matrix and acid treated conductive fillers (ATCBn, ATAB, ATCBe, ATGT) in ratio of 50:25:25

Polymer matrix	Acid treated Conductive filler	Acid treated Conductive filler
50% by weight	25% by weight	25% by weight
PP	ATAB	ATCBn
PP	ATAB	ATGT
PP	ATAB	ATCBe
PP	ATCBn	ATGT
PP	ATCBn	ATCBe
PP	ATGT	ATCBe

*Note: PP is Polypropylene, ATAB is Acid treated Acetylene black, ATCBe is Acid treated Carbon black ENSACO 250, ATAB is Acid treated Carbon black N330, ATGT is Acid treated graphite RGN-C

Part 3 Preparation of polypropylene composite that contain of the optimum ratio of polypropylene composite from part 1 and 2 with CNTs (Timesnano) and CNT (N7000) (Table 3.12)

The optimum ratio of Polypropylene and conductive filler were further investigated by adding mixture of carbon nanotubes of 2%, 3% and 4% by mass as shown in Table 3.12

Table 3.12 Preparation optimum ratio of Polypropylene matrix and acid treated conductive fillers (ATCBe) with carbon nanotubes.

Polymer matrix	Acid treated Conductive filler	Acid treated Conductive filler	Carbon nanotubes	Carbon nanotubes
PP	ATAB	ATCBe	CNTs (N7000)	CNTs (Timesnano)
50% by weight	24% by weight	24% by weight	2% by weight	-
50% by weight	23.5% by weight	23.5% by weight	3% by weight	-
50% by weight	23% by weight	23% by weight	4% by weight	-
50% by weight	24% by weight	24% by weight	-	2% by weight
50% by weight	23.5% by weight	23.5% by weight	-	3% by weight
50% by weight	23% by weight	23% by weight	-	4% by weight

*Note: PP is Polypropylene, ATAB is Acid treated Acetylene black, ATCBe is Acid treated Carbon black ENSACO 250, CNTs (N7000) is Carbon nanotubes N7000, CNTs (Timesnano) is Carbon nanotubes (Timesnano)

3.5 Composite molding

3.5.1 Blending of various ratio of Polypropylene (PP) matrix and conductive fillers by two-roll mill

Polymer composite was blended by two-roll mill. The temperature was at 190°C (Figure 3.1). The preheat time was about 30 minutes and then adjusted the gap between two roll for enhancing shear. After polypropylene was added into the roll and the additives (Carbon black N330 (CBn), Acetylene black (AB), Graphite RGN-C (GT), Carbon black ENSACO 250 (CBe), Carbon nanotubes (CNTs), acid treated carbon black N330 (ATCBn), acid treated acetylene black (ATAB), acid treated graphite RGN-C (ATGT) and acid treated carbon black ENSACO 250 (ATCBe) was further added with adjusting the gap between roll for optimum distance, mix it for 45 minutes (Figure 3.1). Let it cool down and grind it with grinding machine (Figure 3.2).

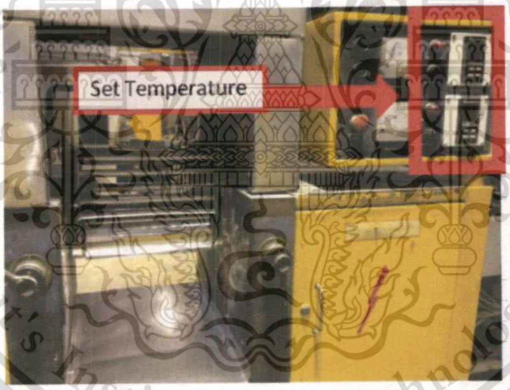


Figure 3.1 Two roll mill (temperature setting)

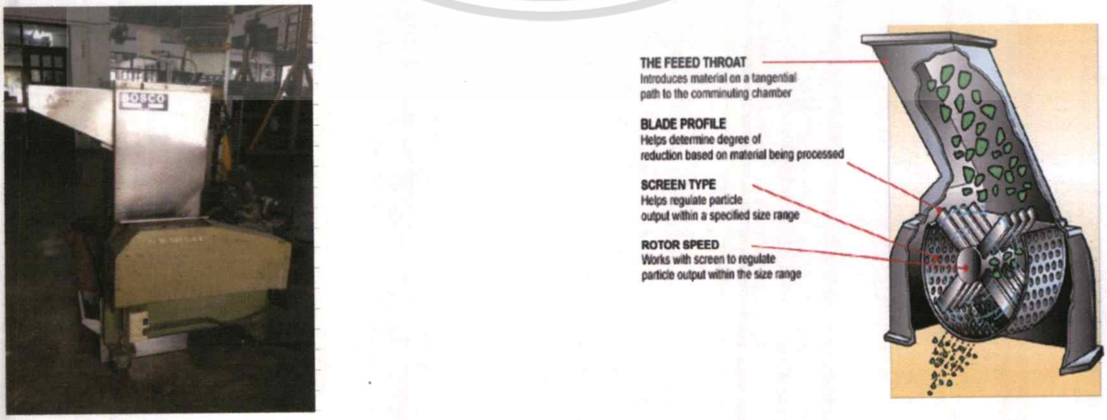


Figure 3.2 grinding machine

3.5.2 Compression of Polypropylene (PP) matrix and conductive fillers by compression molding

The ground composites materials were pressed at pressure of 100 MPa, temperature of 190°C for 6 minutes. After that puts the sample into molding. Let it heating for pressure of 100 MPa and 6 minute and followed by cooling at 100 MPa and 6 minutes.

Figure 3.3 Compression molding

3.6 Characterization of composite

3.6.1 Electrical properties

3.6.1.1 Electrical conductivity by in-plane technique

The sample was prepared with the size of 2 cm x 12 cm x 0.2 cm and then was dried in an oven at 100 °C for 2 hour. Decrease contact resistivity by Silver paint. Give current to 1 and 4 point and measure voltage of 2 and 3 (Figure 3.4). And then, calculate the electrical conductivity followed equation (3.1, 3.2, and 3.3).

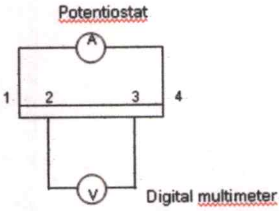


Figure3.4 Four-point probe technique

$$R = \frac{V}{I} \tag{3.1}$$

$$\rho = \frac{R*A}{L} \tag{3.2}$$

$$\sigma = \frac{1}{\rho} \tag{3.3}$$

Where;

ρ = electrical resistivity (Ω.cm)

σ = conductivity (S.cm^{-1})

V = electrical potential (V)

R = electrical resistance (Ω)

I = electric current (A)

A = cross-sectional area (cm^{2})

L = length between point 2 and 3 (cm)

3.6.1.2 Electrical conductivity by Through-plane technique

Electrical conductivity in through-plane of sample was measured followed by Figure 3.5

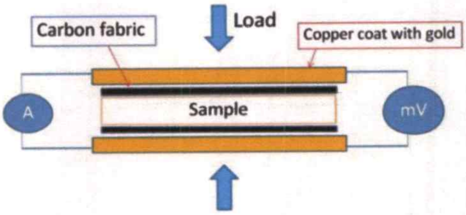


Figure 3.5 Apparatus used for measure resistivity in through-plane of sample [34]

Conductivity in through-plane can prepare anode and cathode by using copper coat with gold plate and carbon fabric plate. Prepare sample that impact to carbon fabric followed by 1 cm.

x 2 cm. x 0.2 cm. and then give load to minimize gap between sample, carbon fabric and copper coat with gold plate. And then, give current in range of 0.1 – 0.5 Ampere and measure voltage of sample by multimeter. Plot graph between voltage and current to calculate resistivity from slope of graph. And calculate electrical conductivity of sample. And then, calculate the electrical conductivity followed equation (3.1, 3.2, 3.3 and 3.4)

$$\mathrm{ICR} = \frac{R_1 - R_2}{2} \times A \qquad (3.4)$$

Where;

R1 = electrical resistance of sample

R2 = electrical resistance of apparatus

3.6.1.3 Resistivity by Through-plane technique

Resistivity in through-plane of sample was measured followed by Figure 3.5 Resistivity in through-plane can prepare anode and cathode by using copper coat with gold plate and carbon fabric plate. Prepare sample that impact to carbon fabric followed by 1 cm. * 2 cm. * 0.2 cm. and then give load to minimize gap between sample, carbon fabric and copper coat with gold plate. And then, give current in range of 0.1 – 0.5 Ampere and measure voltage of sample by multimeter. Plot graph between voltage and current to calculate resistivity from slope of graph. And calculate Interfacial Contact Resistance (ICR) of sample.

3.6.2 Physical properties

3.6.2.1 Density

The sample (2 cm. x 4 cm. x 0.3cm) was weight by balance to measure mass of sample.

And then, calculate the density by equation (3.5)

Calculate: Density = mass / volume (3.5)

3.6.2.2 Water absorption

The sample was prepared with ASTM D570 method. For removing moisture content, the sample from compression molding was dried in an oven for about 1 hour at 100 °C. After that, sample was marked as dry weight. Put dry sample into water for 24 hour and marked as wet weight. Calculate water absorption followed equation (3.6)

Water absorption = [(wet weight – dry weight)/ dry weight] x100 (3.6)

3.6.3 Mechanical Properties

3.6.3.1 Hardness

The sample was prepared by 12 cm x 12 cm x 0.3 cm and then hardness was tested by Durometer shore D (ASTM D2240). Test for 5 point on the sample and then find an average (Figure 3.7). Durometer is a hardness scale based on the indentation hardness of a material. The durometer test determines the hardness by measuring the depth of penetration of an indenter under a large load compared to the penetration made by a preload.

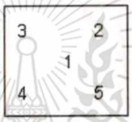


Figure 3.6 five points on sample for hardness

3.6.3.2 Flexural strength

To study flexural strength from ASTM D790-03 [35]. Flexural strength will test at 23°C. Prepare polymer composites in length about 120 mm, width about 10 mm. and high about 3 mm. Rate that use in test is 10 mm./minute. Span length is 50 mm. Test it 5 samples and measure the value of flexural strength.

3.6.4 Morphology by Scanning Electron Microscope (SEM)

Morphology was measured by Scanning Electron Microscope (SEM). Samples are coated with a very thin layer of gold by using a machine called a sputter coater. The sample is placed inside the microscope's vacuum column through an air-tight door.

Figure3.7 Scanning Electron Microscope (SEM) [36]

Chapter 4

Results and discussion

Bipolar plate can be made from various materials with the most common being metal, graphite, and polymer composites. The key is to investigate the development of sufficient electrical conductivity and mechanical properties of polypropylene composite for rapid continuous manufacturing. The effect of the polypropylene with different types and amount of fillers (AB, CBn, GT, CBe, ATAB, ATCB, ATGT, ATCBe and CNTs (Timesnano and N7000) on the electrical conductivity, density, hardness, water absorption and flexural strength of the composite would be analyzed. The results of the experiment can be divided into three parts. First part was prepared polypropylene composite that contain of PP matrix and conductive fillers (CBn,AB,GT,CBe) in ratio of 50:50 and 50:25:25 as shown in Table 3.8-3.9. Second part was prepared polypropylene composite that contain of PP matrix and acid-treated conductive fillers (ATCBn,ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25 (Table 3.10 – 3.11) Third part was prepared polypropylene composite that contain of the optimum ratio of polypropylene composite from part 1 and 2 with CNTs(Timesnano) and CNT(N7000) (Table 3.12)

4.1 Study effect of polypropylene composite that contain of polypropylene matrix and conductive fillers (CBn,AB,GT,CBe) in ratio of 50:50 and 50:25:25(Table 3.8-3.9).

4.1.1 Electrical properties

4.1.1.1 Electrical conductivity by in-plane technique

Electrical conductivity by in-plane technique of Polypropylene matrix and conductive fillers (CBn, AB,GT,CBe) in ratio of 50:50 and 50:25:25 were shown in Figure 4.1

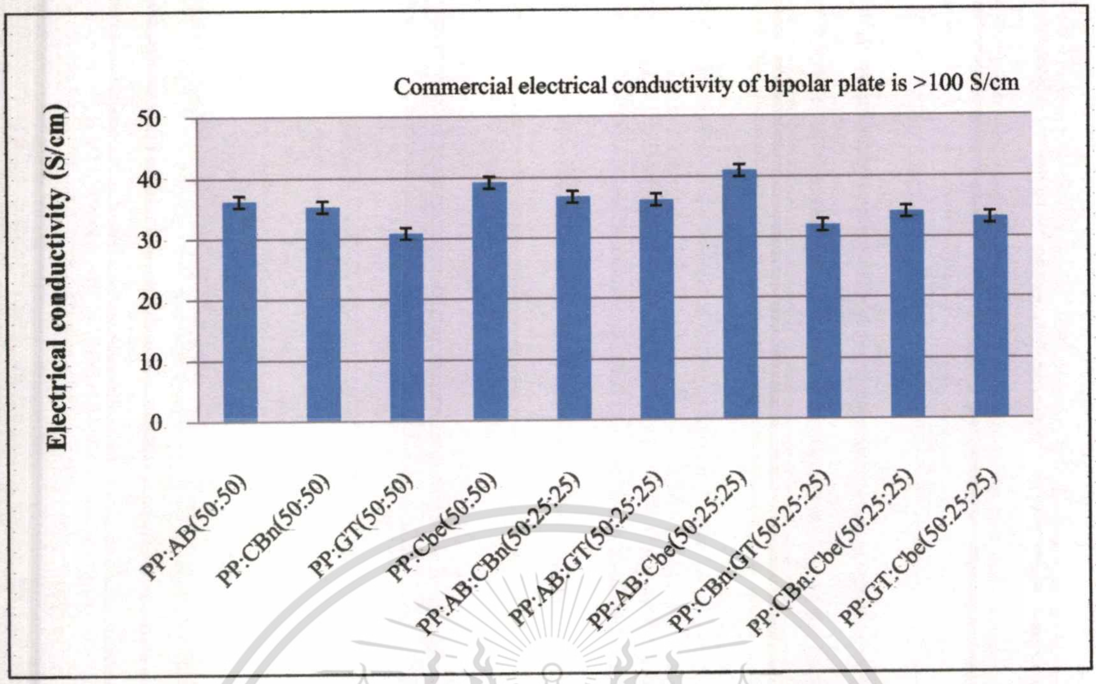


Figure 4.1 Electrical conductivity by in-plane technique of Polypropylene matrix and conductive fillers (CBn, AB,GT,CBe) in ratio of 50:50 and 50:25:25

Figure 4.1 shows the electrical conductivity of Polypropylene matrix and conductive fillers (CBn,AB,GT,CBe) in ratio of 50:50 and 50:25:25 was in the range of 30.89 – 40.93 S/cm. (Appendix B-1) The lowest electrical conductivity of ratio 50:50 (30.89 S/cm) was PP: GT (50:50). This according to GT filler had the lowest purity about 82.6% compared to other fillers. The highest electrical conductivity of PP matrix and conductive fillers in ratio of 50:50 (39.22 S/cm) was PP:CBe(50:50), it was indicated the highest electrical conductivity because CBe has the highest purity of 99.79% and had a smaller particle size of 0.04 nm., therefore, CBe could be penetrated inside PP matrix better than other fillers. When two types of fillers were mixed into PP matrix (CBn,AB,GT,CBe) in ratio of 50:25:25, the electrical conductivity was in range of 32.02 – 40.93 S/cm. The lowest electrical conductivity of PP matrix and conductive fillers in ratio of about 50:25:25 (32.02 S/cm.) was PP:CBn:GT (50:25:25). The highest electrical conductivity of PP matrix and conductive fillers in ratio of 50:25:25 (40.93 S/cm.) was PP:AB:CBe (50:25:25). It was indicated the highest electrical conductivity because both of AB and CBe had higher purity than CB(N330) and RGN-C that they could be affected to the electrical current and pass through those conductive AB and CBe particles instead of only insulating PP matrix. Another reason that they affected to the electrical conductivity of PP:AB:CBe(50:25:25) because a small CBe particles filled up the big pores among larger AB particle; thus the electrical current can easily move through the polymer composite PP:AB:CBe(50:25:25).

4.1.1.2 Electrical conductivity by through-plane technique

Electrical conductivity by through-plane technique of Polypropylene matrix and conductive fillers (CBn, AB,GT,CBe) in ratio of 50:50 and 50:25:25 were shown in Figure 4.2

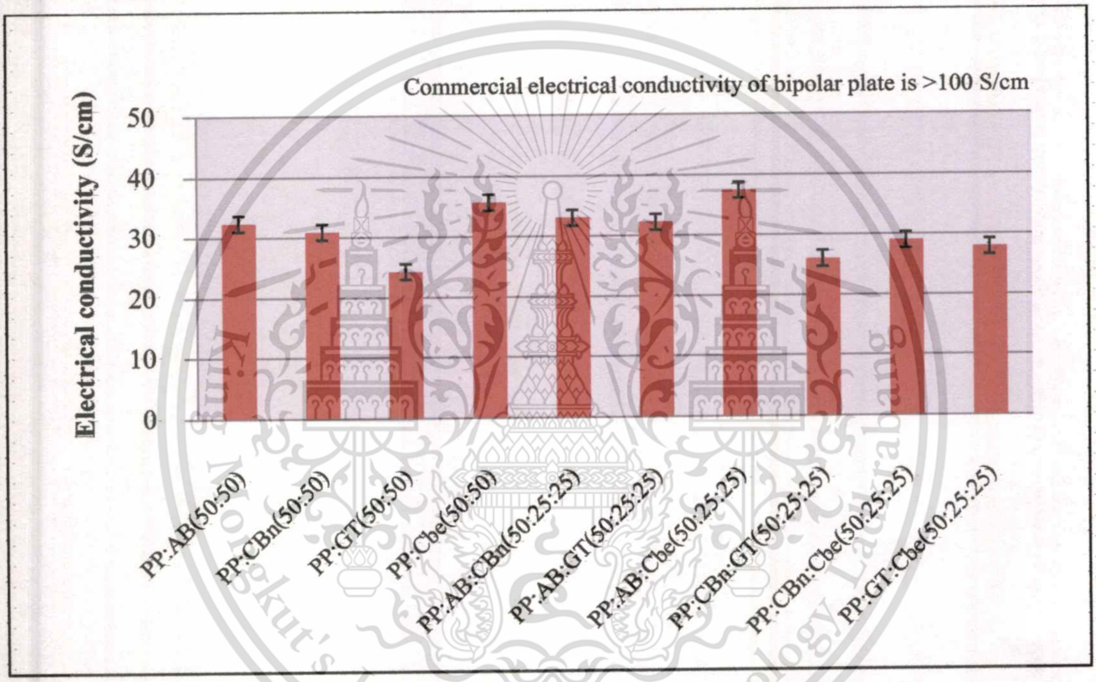


Figure 4.2 Electrical conductivity by through-plane technique of Polypropylene matrix and conductive fillers (CBn,AB,GT,CBe) in ratio of 50:50 and 50:25:25

Electrical conductivity by through-plane technique of Polypropylene matrix and conducting fillers (CBn,AB,GT,CBe) in ratio of 50:50 and 50:25:25 was in range of 24.29– 37.47 S/cm. (Appendix B-2) The lowest electrical conductivity was about 24.29 S/cm (PP: GT (50:50)), because GT filler had lowest purity of 82.6% compared to other fillers. The highest electrical conductivity of PP matrix and conductive fillers in ratio of 50:50 (35.66 S/cm) was PP:CBe(50:50), This was according to high electrical conductivity of CBe which had the highest purity of 99.79% and had a smaller particle size of 0.04 nm., therefore CBe could be penetrated inside PP matrix better than other fillers. When two types of fillers (CBn, AB, GT, CBe) were mixed into PP matrix. They were polypropylene matrix and conductive fillers (CBn,AB,GT,CBe) in ratio of 50:25:25, the electrical conductivity was in range of 26.03 – 37.47 S/cm. The highest electrical conductivity of PP matrix and conductive fillers in ratio of 50:25:25 (37.47 S/cm.) was PP:AB:CBe (50:25:25). It was dominated the highest electrical conductivity because both AB and CBe had higher purity than CB (N330) and RGN-C that affected to the electrical current. They could be passed through those conductive AB and CBe particles instead of only insulating PP matrix. Another reason affected to the electrical conductivity of PP:AB:CBe(50:25:25) was a small CBe particles filled up the big pores among larger AB particle; thus the electrical current could easily move through the polymer composite PP:AB:CBe(50:25:25). The through-plane electrical conductivity of the composite was lower than the in-plane electrical conductivity because the particles of fillers were randomly distributed within the matrix and aligned in parallel pressure direction of the composite.[37] There were affected to penetration ability of additives into the matrix, the compatibility of fillers and purity of fillers. However, CBe had more significant effect than other fillers.

4.1.1.3 Resistivity by Through-plane technique (Interfacial Contact Resistance (ICR))

Interfacial Contact Resistance (ICR by Through-plane technique of Polypropylene matrix and conductive fillers (CBn, AB,GT,CBe) in ratio of 50:50 and 50:25:25were shown in Figure 4.3

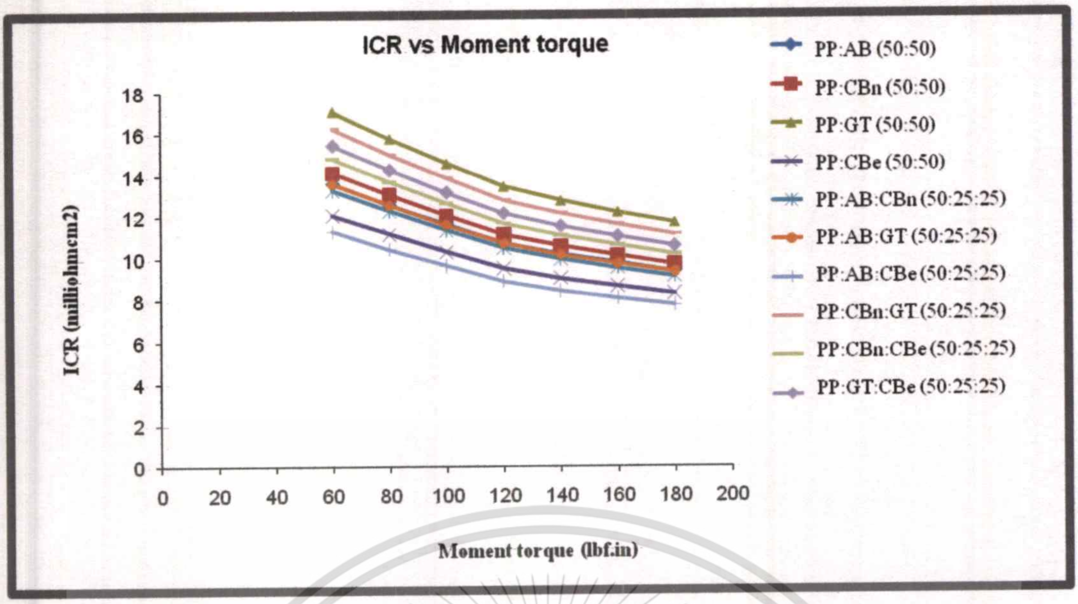


Figure 4.3 Interfacial Contact Resistance by Through-plane technique of Polypropylene matrix and conductive fillers (CBn,AB,GT,CBe) in ratio of 50:50 and 50:25:25.

Figure 4.3 shows the Interfacial Contact Resistance (ICR) of PP that having different ratios of conductive fillers. The Interfacial Contact Resistance (ICR) was decreased when moment torque was increased to the sample because electron could be easily passed through the composite. The lower the Interfacial contact resistance, the higher the electrical conductivity was. The lowest interfacial contact Resistance was dominated in PP:AB:CBe with ratio of 50:25:25. (Appendix B-3)

4.1.2 Physical properties

4.1.2.1 Density

Volume and weight of Polypropylene and conductive fillers (CBn,AB,GT,CBe) in ratio of 50:50 and 50:25:25 were measured by vernier caliper. Then density was calculated by equation (3.5). The results were shown in Figure 4.4 (Appendix A-4)

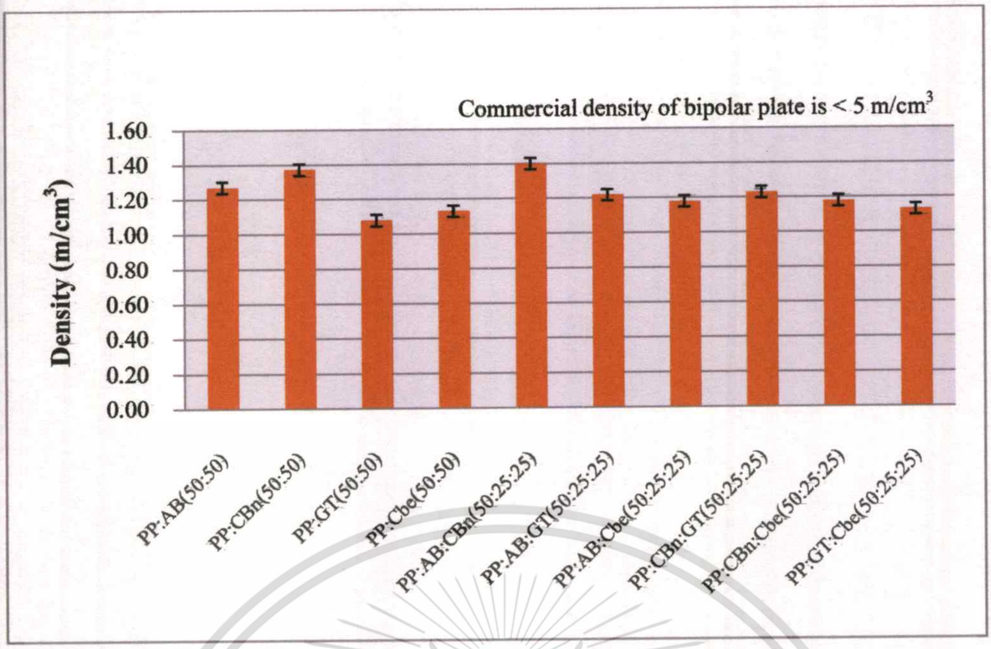


Figure 4.4 Density of Polypropylene matrix and conductive fillers (CBn,AB,GT,CBe) in ratio of 50:50 and 25:25

Figure 4.4 shows the density of Polypropylene and conductive fillers (CBn,AB,GT,CBe) in ratio of 50:50 and 50:25:25 (1.08-1.40 g/cm^3) (Appendix B-4) was higher than the density of PP itself (0.90 g/cm^3). The densities of composites were increased with filler concentration. The composite that contain CBn (Carbon black N330) has more density than other because the density of CBn filler itself was about 0.320 g/cm^3 that was more than the density of AB and CBe but it was closed to density of GT. This was depending on compatability of fillers and matrix. The lowest density (1.08 g/cm^3) was PP: GT (50:50) because the highest density (1.40 g/cm^3) was PP:AB:CBn in ratio of 50:25:25. This was according to the synergistic effect between fillers and matrix which caused by the penetration ability of fillers into PP matrix. [38] The density of composite was less than 5 g/cm^2 which was specified in commercial grade. Lower density is a key component, as a bipolar plate needs to be light and thin in order to reduce the overall cost.

4.1.2.2 Water absorption of polypropylene composite followed by ASTM D 570

The samples from compression molding were dried in an oven for removing moisture content about 24 hour at 80°C. After that, sample was marked as dry weight. Dry sample was put into water for 2 hours and marked as wet weight. Calculated water absorption followed by equation (3.6). The results were shown in Figure 4.5

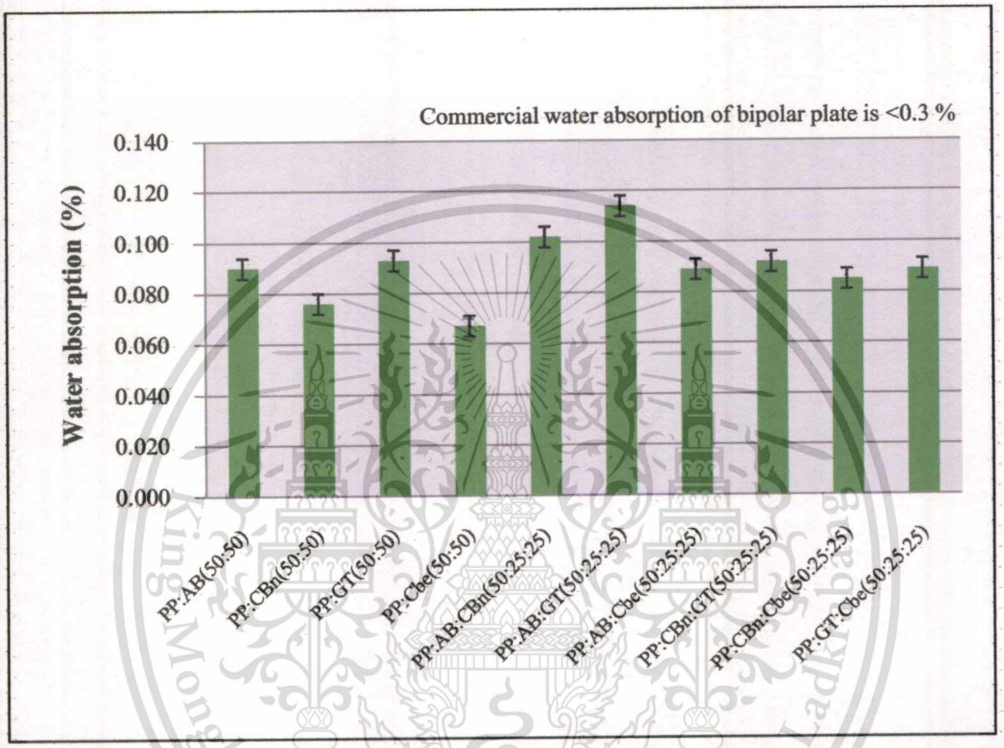


Figure 4.5 Water absorption of Polypropylene and conductives fillers (CBn, AB,GT,CBe) in ratio of 50:50 and 50:25:25

Figure 4.5 shows the water absorption of composite had the same trend. The water absorption of composite was in range of 0.067 - 0.114 %. (Appendix B-5) The highest water absorption (0.114%) was PP:AB:GT (50:25:25). It was showed the lowest water absorption (0.067%) that implied the good performance of bipolar plate. The water absorption of composite was less than 0.3%, therefore it was specified in commercial grade. However, the water absorption in this research was no significant concern. This was according to observed value that between layers of polypropylene composites, there are low empty spaces. So, it could absorp less amount of water. These spaces are due to the pressure applied during compression moulding.

4.1.3 Mechanical Properties

4.1.3.1 Hardness test

The samples were prepared from compression molding. Shore D (HRD) was use to identify hardness of Polypropylene with conductive fillers (CBn,AB,GT,CBe) in ratio of 50:50 and 50:25:25 followed by ASTM D2240. There were shown in Figure 4.6

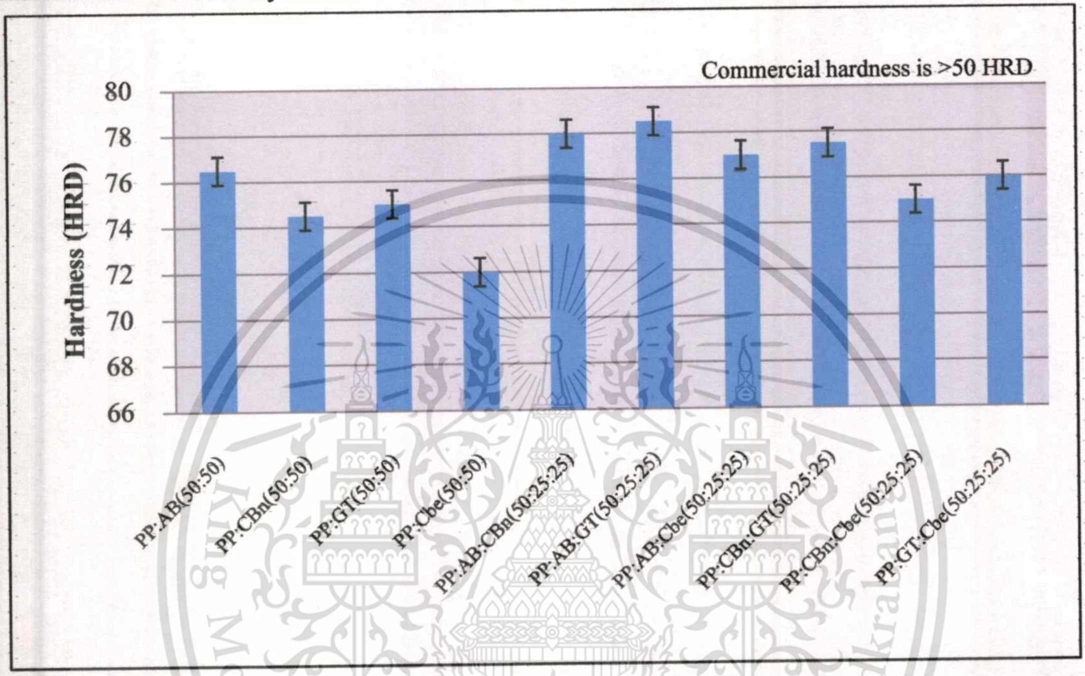


Figure 4.6 Hardness of Polypropylene and conductives fillers (CBn,AB,GT,CBe) in ratio of 50:50 and 50:25:25

Figure 4.6, the hardness of composite was 72 - 78.5 HRD in range. (Appendix B-6) The hardness of PP was not much change when fillers (CBn,AB,GT,CBe) were added. The highest hardness was 78.5 HRD received from 50%wt of PP, 25%wt of AB and 25%wt of GT. The lowest hardness was (72 HRD) PP:CBe(50:50). The hardness of PP with AB was more than CBn, GT, and CBe because of AB had larger particle size than the other fillers. Therefore, this was not reduced spaces between filler and matrix. The hardness was increased because the hardness of composites relies on the matrix. This was due to penetration ability of fillers seated into the matrix PP. However, there are not much changes in hardness of PP and conductives fillers

(CBn,AB,GT,CBe) composite. Therefore, the hardness of composites was passed through the commercial requirement (> 50 HRD).

4.1.3.2 Flexural strength

The flexural strength of composite plate was measured by three point bending test using a Universal Testing Machine (LLOYD LR 10K PLUS) as followed ASTM standard D790. Rectangular polypropylene composites of height 10 mm., width of 3 mm. and length of 120 mm. in dimensions were placed on two supports with span length 50 mm and rate about 10 mm./minute. A load at the midpoint of the samples was initiated in the way that was presented in Figure 4.7

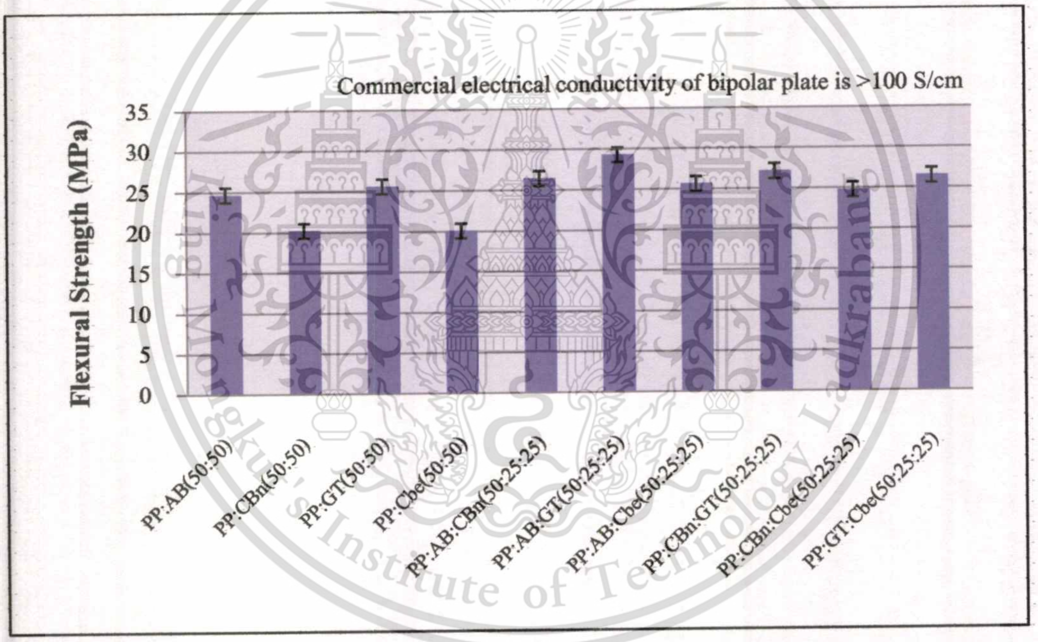


Figure 4.7 The flexural strength of the polypropylene with conductive fillers (CBn,AB,GT,CBe) in ratio of 50:50 and 50:25:25

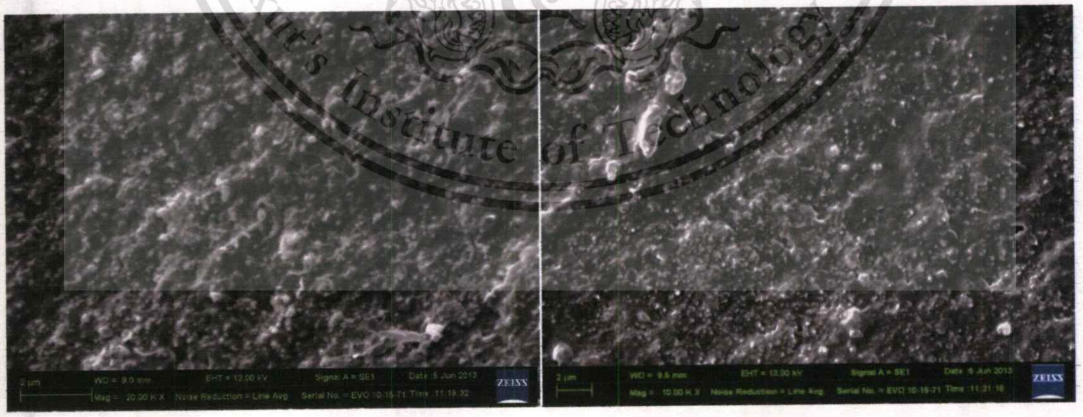
Figure 4.7 shows that flexural strength varied with different fillers. A similar result was obtained in the case of adding fillers (CBn,AB,GT,CBe). The measured flexural strength for the polymer composites bipolar plates were in range of 20.09 – 29.25 MPa. (Appendix B-7) The results indicated that the flexural strength had the same trend. The addition of 25%wt AB and 25%wt GT was confirmed the greatest effect of 29.25 MPa. The lowest flexural strength (20.09

MPa was PP:CBe(50:50). However, the polymer composite doped with high filler loadings were difficult to reach high flexural strengths of composite bipolar plate. The results were still below the commercial flexural strength (>59 MPa). This was according to the agglomeration and poor bonding of PP with conductive fillers in composite.

4.1.4 Morphology by scanning Electron Microscope (SEM)

A Scanning Electron Microscope (SEM) technique was an effective way to view the microstructure of composite for explanation dispersability of polypropylene with conductive fillers (CBn,AB,GT,CBe) in ratio of 50:50 and 50:25:25. The results were shown in Figure 4.8-4.9

a.) PP:AB (50:50) b.) PP:CBn (50:50)

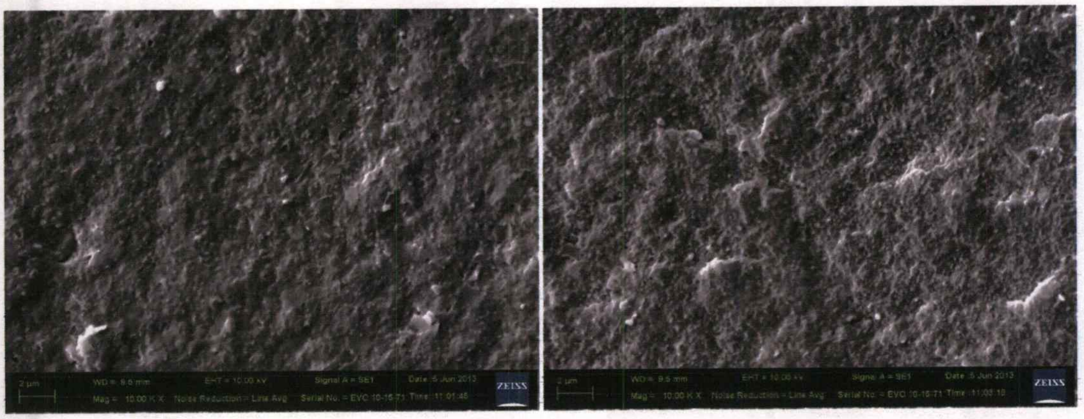


c.) PP:GT(50:50) d.) PP:CBe(50:50)

Figure 4.8, Morphology of polypropylene with conductive fillers (CBn,AB,GT,CBe) in ratio of 50:50 with magnification 10,000 times. a.) PP:AB(50:50) b.) PP:CBn(50:50) c.) PP:GT(50:50) d.) PP:Cbe(50:50)

Figure 4.8 shows morphology of polypropylene with conductive fillers (CBn,AB,GT,CBe) in ratio of 50:50 with magnification 10,000 times. In this technique the dispersability of composites were studied. The optimum of superior dispersability was polymer composite in Figure 4.8(d.) when it was compared with the other. CBe had smaller of particle size than other. Therefore, it can mix with polymer matrix better than other filler. The morphology of polymer composite was observed that between layers of polypropylene composites, there is low empty space. Therefore, water absorption of composite was less than 0.3% (commercial grade). Polypropylene composite in Figure 4.8 (a.) shows more space between filler and matrix than the other so, the hardness was increase too because it was relies on the matrix. From the morphology of composite in Figure 4.8 a,b,c and d was dominated the agglomeration and poor bonding of PP with conductive fillers. Therefore, the results were still below the commercial flexural strength (>59 MPa).

a.) PP:CBe (50:50) b.) PP:CBe:AB (50:25:25)



c.) PP:CBe:CBn (50:25:25)

d.) PP:CBe:GT:CBe (50:25:25)

Figure 4.9, Morphology of polypropylene with conductive fillers (CBn,AB,GT,CBe) in ratio of 50:25:25 with magnification 10,000 times. a.) PP:CBe (50:50) b.) PP:CBe:AB (50:25:25) c.) PP:CBe:CBn (50:25:25) d.) PPLCBe:GT:CBe

Figure 4.9 shows the morphology of the best optimum ratio of polypropylene composite with one filler in ratio of PP:filler (50:50) compared with poly propylene composite that contain of two types of fillers in ratio of PP:filler1:filler2 (50:25:25) with magnification 10.000 times. In this technique were studied the dispersability of composite. The optimum of superior dispersability was polypropylene composite in Figure 4.9 (b.) (PP:CBe:AB in ratio of 50:25:25) that was more electrical conductivity than polypropylene composite that have one type of filler (Figure 4.9 (a.) (PP:CBe in ratio of 50:50)) because particles of filler contact one another, a continuous path was formed through the polymer matrix, for electrons to travel through. From the morphology of composite, there are low empty spaces between layers of polypropylene composite (Figure 4.9 a,b,c and d). So, it could absorp less amount of water. Therefore, water absorption of composite was passed commercial grade (commercial water absorption is less than 0.3%).

4.2 Study effect of polypropylene composite that contain of Polypropylene matrix and acid-treated conductive fillers (ATCBn, ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25.

4.2.1 Electrical properties

4.2.1.1 Electrical conductivity by in-plane technique

The electrical conductivity by in-plane technique of polypropylene matrix and acid-treated conductive fillers (ATCBn,ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25 were shown in Figure 4.10

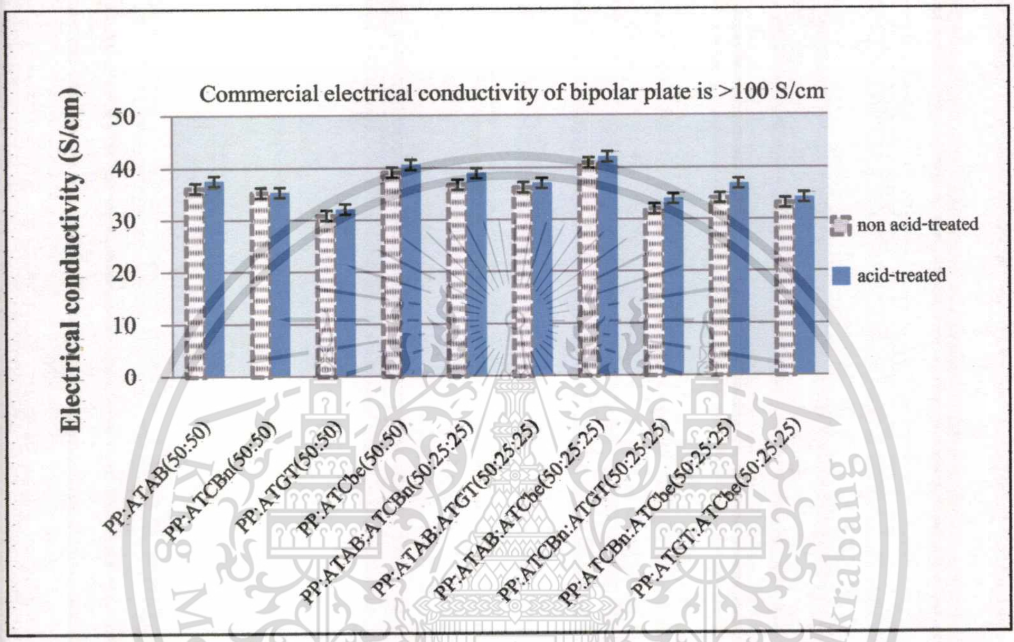


Figure 4.10 Electrical conductivity by in-plane technique of Polypropylene matrix and acid-treated conductive fillers (ATCBn, ATAB, ATGT, ATCBe) in ratio of 50:50 and 50:25:25

Figure 4.10 shows electrical conductivity of Polypropylene matrix and acid-treated conducting fillers (ATCBn,ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25 in range of 32.12 – 42.06 S/cm. (Appendix C-1) The electrical conductivity was higher when it was compared to PP with non acid-treated fillers composite in part 4.1. When the conductive filler were treated with acid, there were more purity and more porosity. Therefore, the particle of acid-treated fillers could be attached to each other more than particle of non-acid treated fillers. This was affected to the movement of electrical current. Then, electrical current could be continuously moved through the composite; hence the electrical conductivity was increased. The lowest electrical conductivity (32.12 S/cm) was PP: GT in ratio of 50:50, because GT filler had the lowest purity of 82.6% compared to other fillers. The highest electrical conductivity (40.65 S/cm) of PP matrix and conductive fillers in ratio of 50:50 was PP:ATCBe(50:50), it dominated the high electrical conductivity because ATCBe possessed the highest purity of 99.79% and had a smaller particle size of 0.04 nm., therefore ATCBe could be penetrated inside of PP matrix better than other fillers. [39] When two types of fillers were mixed (ATCBn,ATAB,ATGT,ATCBe) into PP matrix in ratio of 50:25:25, the electrical conductivity was in range of 33.98 – 42.07 S/cm. The highest electrical conductivity (42.07 S/cm) of PP matrix and conductive fillers in ratio of 50:25:25 was PP:ATAB:ATCBe (50:25:25). Because it dominated the highest electrical conductivity according to both ATAB and ATCBe possessed higher purity more than ATCB(N330) and RGN-C which affected to the electrical current passing through those conductive ATAB and ATCBe particles instead of insulating PP matrix. [39] The other reason of PP:ATAB:ATCBe(50:25:25) was a small ATCBe particles filled up the big pores among larger ATAB particle; thus the electrical current could easily move through the polymer composite PP:ATAB:ATCBe(50:25:25). The increasing amount CBe (Carbon Ensaco 250) was enhanced the higher electrical conductivity according to the effect of penetration ability of additives into the matrix, structure and purity of fillers.

4.2.1.2 The electrical conductivity by through-plane technique

The electrical conductivity by through-plane technique of polypropylene matrix and acid – treated conductive fillers (ATCBn,ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25 were shown in Figure 4.11

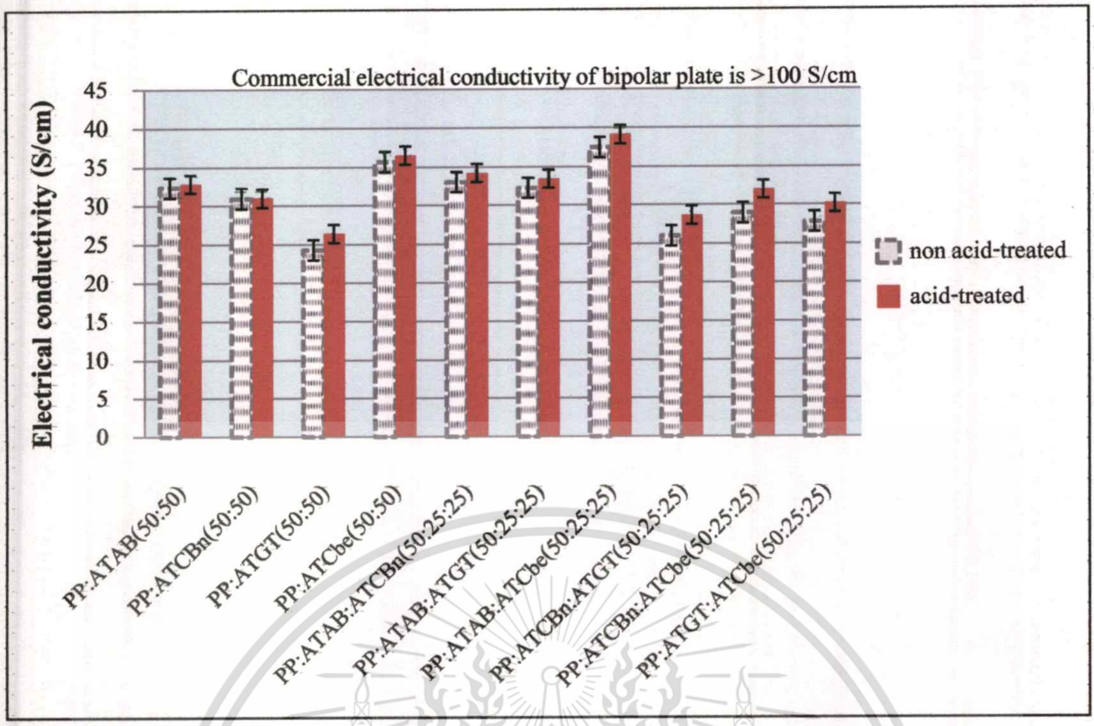


Figure 4.11 Electrical conductivity by through-plane technique of Polypropylene matrix and conductive fillers (ATCBn,ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25.

Figure 4.11 shows electrical conductivity of Polypropylene matrix and acid-treated conductive fillers (ATCBn,ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25 in range of 26.40 – 39.12 S/cm. (Appendix C-2) The electrical conductivity was higher when it was compared to PP with non acid-treated fillers composite in part 4.1. When the conductive fillers were treated with acid, there were more purity and more porosity. Therefore, the particle of acid treated fillers could be attached to each other more than particle of non acid-treated fillers. This was affected to the movement of electrical current. Then, electrical current could be continuously moved through the composite; hence the electrical conductivity was increased. The lowest electrical conductivity (26.40 S/cm) was PP:GT in ratio of 50:50 because GT filler had the lowest purity of 82.6% compared to other fillers. The highest electrical conductivity (36.49 S/cm) of PP matrix and conductive fillers in ratio of 50:50 was PP:ATCBe), It dominated the high electrical conductivity because ATCBe possessed the highest purity of 99.79% and had a smaller particle size of 0.04 nm., therefore ATCBe could be penetrated inside of PP matrix better than other fillers. And after treated by acid, ATCBe had a higher surface area of 98.85 m^2/g, therefore ATCBe could mix with matrix better than non-acid treated CB. When two types of fillers (ATCBn,ATAB,ATGT,ATCBe) were mixed into PP matrix in ratio of 50:25:25, the electrical conductivity was in range of 28.68 – 39.12 S/cm. The highest electrical conductivity (39.12 S/cm) of PP matrix and conductive fillers in ratio of 50:25:25 was PP:ATAB:ATCBe (50:25:25). Because it dominated the highest electrical conductivity according to both ATAB and ATCBe possessed higher purity more than ATCB (N330) and RGN-C which affected to the electrical current passing through those conductive ATAB and ATCBe particles instead of the insulating PP matrix. The other reson of PP:ATAB:ATCBe (50:25:25) was a small ATCBe particles filled up the big pores among larger ATAB particle, thus the electrical current could easily move through the polymer composite PP:ATAB:ATCBe (50:25:25). The increasing of CBe (Carbon Ensaco 250) was enhanced the higher of the electrical conductivity according to the effect of penetration ability of additives into the matrix, structure and purity of fillers. The through-plane electrical conductivity of the composite was lower than the in-plane electrical conductivity due to the particle of fillers are randomly distributed within the matrix and align in parallel pressure direction of the composite.

4.2.1.3 Resistivity by Through-plane technique

Interfacial Contact Resistance (ICR by Through-plane technique of Polypropylene matrix and acid-treated conductive fillers (ATCBn,ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25 were shown in Figure 4.12

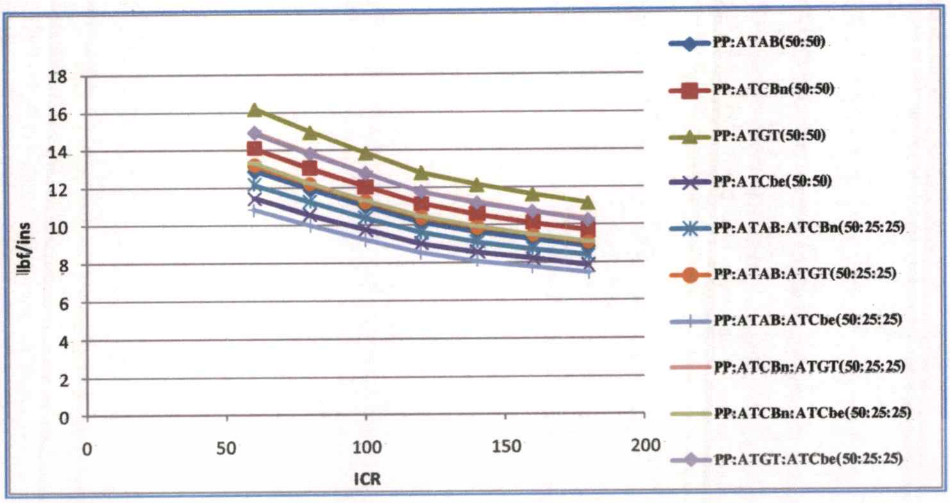


Figure 4.12 Interfacial Contact Resistance by Through-plane technique of Polypropylene matrix and conductive fillers (CBn,AB,GT,ATCBe) in ratio of 50:50 and 50:25:25.

Figure 4.12 shows the Interfacial Contact Resistance (ICR) of PP having different ratios of conductive fillers. The Interfacial Contact Resistance (ICR) was decreased when they increased the moment torque to the sample. The lower the Interfacial, the higher the electrical conductivity was. The lowest interfacial contact Resistance dominated in PP:AB:CBe in ratio of 50:25:25. The interfacial contact resistance was lower when it was compared to PP with non-acid treated filler composite. The particle of acid treated fillers could be attached to each other more than particle of non-acid treated fillers because there were more purity and more porosity than acid treated fillers. (Appendix C-3)

4.2.2 Physical properties

4.2.2.1 Density

Volume and weight of Polypropylene and conductive fillers (ATCBn,ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25 were measured by venire caliper and then calculated followed by equation (3.5). The results were shown in Figure 4.13

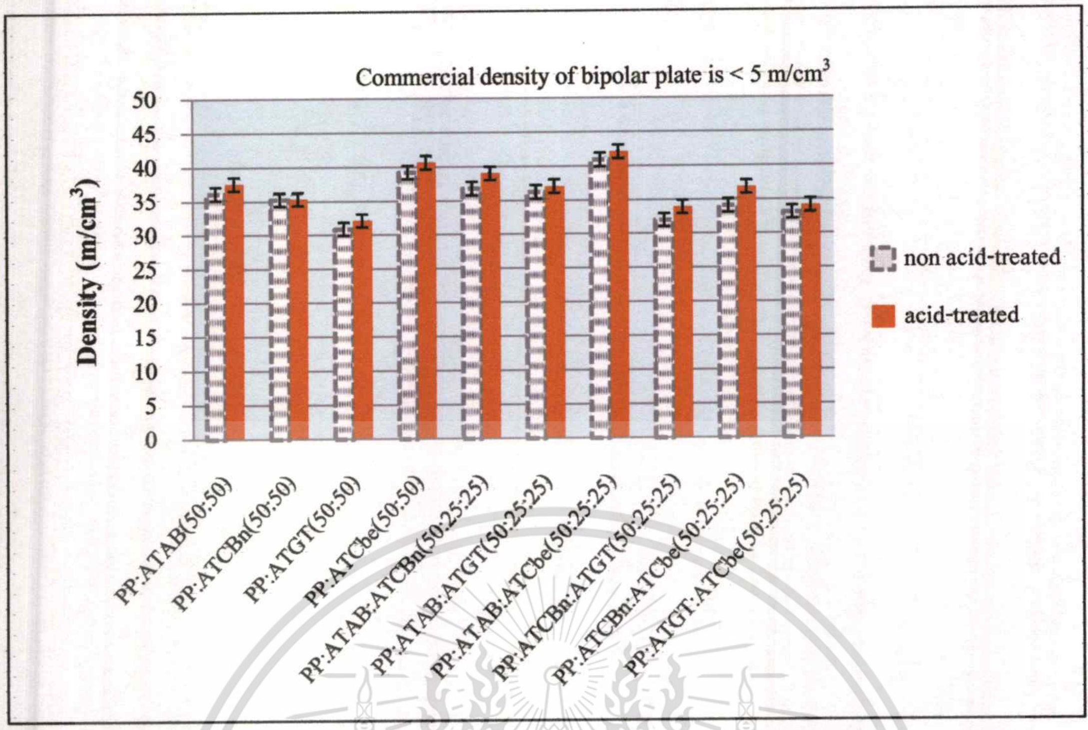


Figure 4.13 Density of Polypropylene and acid-treated conductives fillers (ATCBn, ATAB, ATGT, ATCBe) in ratio of 50:50 and 50:25:25.

The density of Polypropylene and conductive fillers (ATCBn,ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25 (1.1 – 1.47 g/cm^3) (Appendix C-4) was higher than the density of PP itself (0.90 g/cm^3) as shown in Figure 4.13. The densities of composites were increased with filler concentration. The composite that contain of CBn (Carbon black N330) has more density than others because the density of CBn filler itself was about 0.320 g/cm^3 which was more than the density of AB and CBe but it was close to the density of GT. Therefore, it was depend on compatability of fillers and matrix. The lowest density (1.1 g/cm^3) was PP:ATGT:ATCBe (50:25:25). The highest density (1.47 g/cm^3) was PP:ATAB:ATCBn in ratio of 50:25:25 which it was indicated the highest density due to the synergistic effect between fillers and matrix. This was caused by the penetration ability of fillers into PP matrix. The density of PP matrix mixed with acid treated was no change when compared to PP matrix mixed with non-acid treated composite. The density of composite was less than 5 g/cm^3 which was specified in commercial grade. From a manufacturing perspective, the bipolar plate should have low density in order to reduce the overall cost.

4.2.2.2 Water absorption of sample with ASTM D 570

The samples from compression molding were dried in an oven for removing moisture content for 24 hour at 80°C. After that, sample was marked as dry weight. Dry sample was put into water for 2 hours and marked as wet weight. Calculated water absorption followed by equation (3.6). The results were shown in Figure 4.14

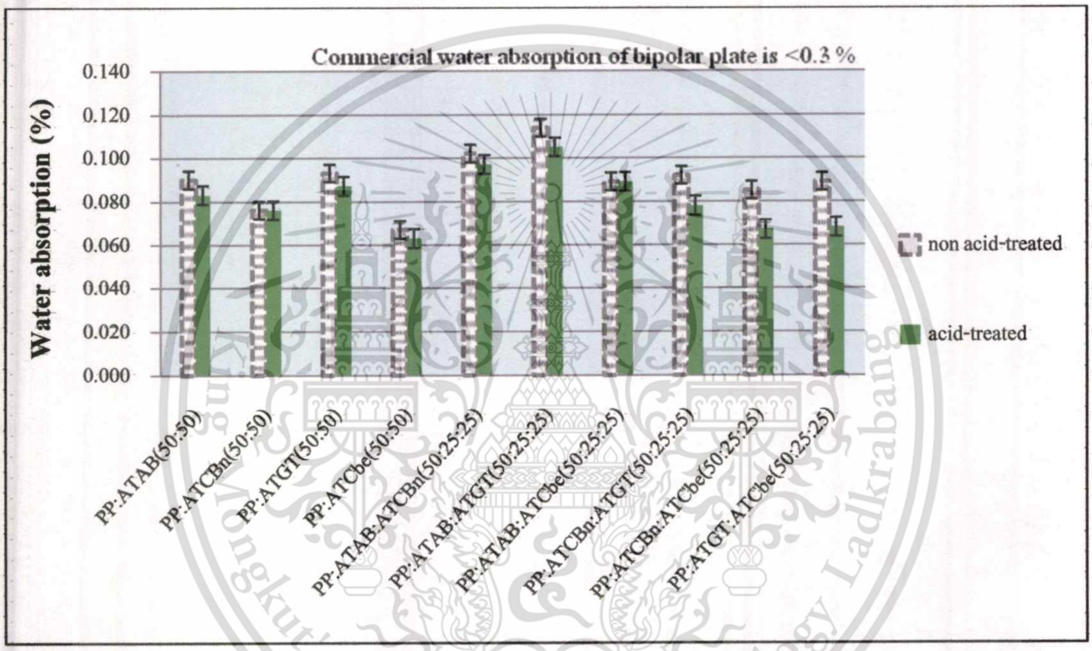


Figure 4.14 Water absorption of Polypropylene and conductives fillers (ATCBn,ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25

The water absorption of composite had the same trend. The water absorption of composite was in range of 0.067 - 0.114 %. (Appendix C-5) The highest water absorption (0.105%) was PP:ATAB:ATGT (50:25:25). PP:ATCBe (50:50) had the lowest water absorption about 0.063% which it was good performance for bipolar plate. However, the water absorption in this research was no change when compared to PP composite that contain of non-acid treated fillers. Therefore, water absorption of composite was less than 0.3% which it was accepted in commercial use. This was according to there are very low spaces between layer. These spaces or gaps are as a result of the pressure applied during compression molding.

4.2.3 Mechanical Properties

4.2.3.1 Hardness test

The samples were prepared from compression molding. Shore D (HRD) was used for testing hardness of Polypropylene with acid-treated conductives fillers(ATCBn,ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25 samples (ASTM D2240). The results were revealed in Figure 4.15.

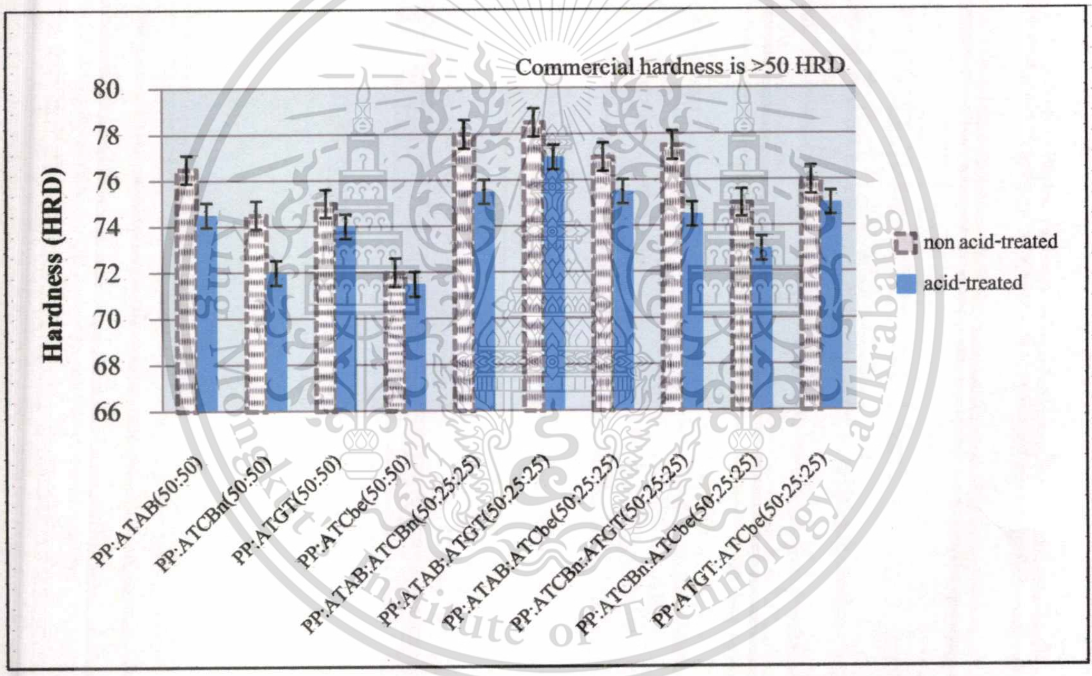


Figure 4.15 Hardness of Polypropylene and acid-treated conductive fillers (ATCBn,ATAB,ATGT, ATCBe) in ratio of 50:50 and 50:25:25

The hardness of composite was 71.5 - 77 HRD in range. (Appendix C-6) The hardness of PP was not much change when fillers (ATCBn,ATAB,ATGT,ATCBe) were added. The highest of hardness (77 HRD) was 50%wt of PP and acid-treated conductive fillers (25% ATAB and 25% ATGT). The lowest of hardness (71.5 HRD) was PP:ATCBe(50:50). The hardness of PP with ATAB was more than ATCBn, ATGT, and ATCBe because ATAB possessed bigger particle size than the other fillers. Therefore it was not decrease the porosity between filler and matrix. The hardness increases because the hardness of composites relies on the matrix. This was due to penetration ability of acetylene black seated into the matrix PP. Shore hardness was decreased when compared to non acid-treated fillers because the acid-treated fillers are very soft and loose porous material than non-acid treated fillers. However, there are no significant changes in hardness of PP and conductive fillers (ATCBn,ATAB,ATGT,ATCBe). Therefore, the hardness of composites was passed through the commercial requirement (> 50 HRD).

4.2.3.2 Flexural strength

The flexural strength of the polypropylene with acid-treated conductive fillers (ATCBn,ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25 were shown in Figure 4.16

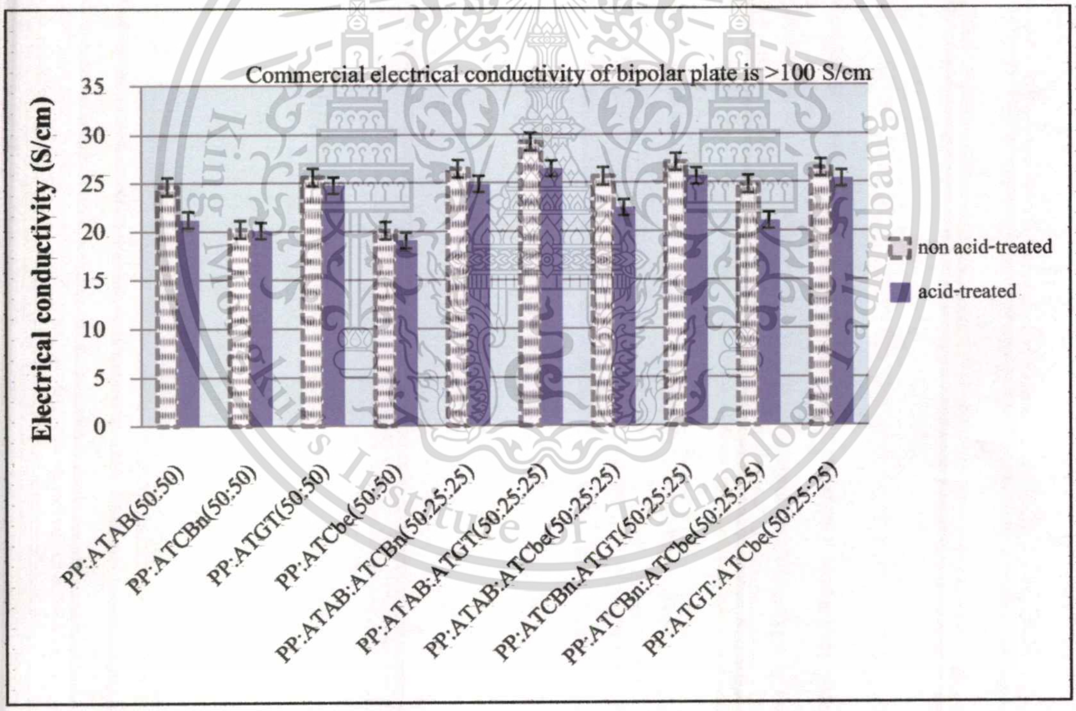
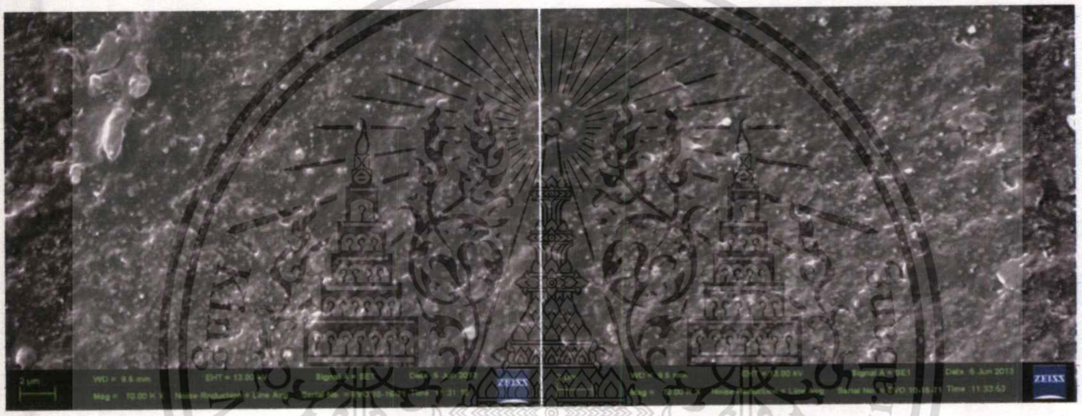


Figure 4.16, Flexural strength of the polypropylene with acid-treated conductive fillers (ATCBn,ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25

Figure 4.16 shows the flexural strength varied with different fillers. A similar result was obtained in the case of adding fillers (ATCBn,ATAB,ATGT,ATCBe). The measured flexural strength for the polymer composites bipolar plates were in range 19.054 – 26.44 MPa. (Appendix C-7) The results were indicated that the same trend of flexural strength of composite. The addition of 25%wt ATAB and 25%wt ATGT into PP matrix were revealed the greatest effect of 26.44 MPa. The lowest flexural strength (19.05 MPa) was PP:CBe(50:50). The results were not corresponding to commercial requirement (> 59 MPa). Flexural strength was decreased when compared to acid-treated fillers because the acid treated fillers are very soft and loose porous material than non-acid treated fillers.

4.2.4 Morphology by Scanning Electron Microscope (SEM)

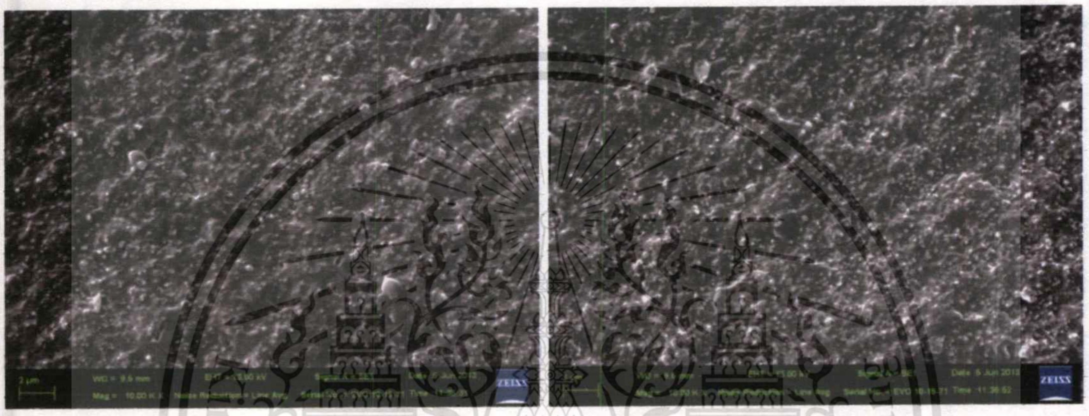


a.) PP:CBe (50:50) b.) PP:ATCBe (50:50)

Figure 4.17 Morphology of the optimum ratio of polypropylene matrix mixed with one type of non acid-treated filler and acid-treated fillers in ratio of 50:50 with magnification 10,000 times. a.) PP:CBe (50:50) b.) PP:ATCBe (50:50)

Figure 4.17 shows the morphology of the optimum ratio of polymer matrix mixed with one type of non-acid treated filler and acid treated filler in ratio of 50:50. The morphology results were shown that the optimum of superior dispersability was PP matrix with acid treated filler that shown in Figure 4.17 (b.) (PP:ATCBe in ratio of 50:50), it was more superior dispersability than non-acid treated filler (Figure 4.17 (a.)) because CBe had more surface area after treated by acid. It had surface area about 65.6 m^2/g (Table 3.5) before treated by acid and then after treated by acid, the filler (CBe) had more porosity and more higher surface area about 98.85 m^2/g (Table3.5)

Therefore, particle of acid treated filler will contact with PP matrix better than non-acid treated filler. This was due to better dispersability of acid treated particle into PP matrix more than non acid-treated filler. This was affected to the movement of electrical current. Then, electrical current could be continuously moved through the composite; hence the electrical conductivity was increase. However, it was the same result to previous work of Saranee [31] that she got higher electrical conductivity of polymer composite that contain of acid-treated filler than polymer composite that contain non-acid treated filler.



a.) PP:CBe:AB (50:25:25) b.) PP:ATCBe:ATAB (50:25:25)

Figure 4.18 Morphology of the optimum ratio of polypropylene matrix mixed with two types of non-acid treated filler and acid-treated fillers in ratio of 50:25:25 with magnification 10,000 times. a.) PP:CBe:AB (50:25:25) b.) PP:ATCBe:ATAB (50:25:25)

Figure 4.18 shows morphology of the optimum ratio of polypropylene composite with two types of non-acid treated filler and acid-treated filler in ratio of 50:25:25 with magnification 10,000 times. This technique were studied the dispersability of composite. The optimum of superior dispersability was polypropylene composite in Figure 4.18 (b.) (PP:ATCBe:ATAB with ratio of 50:25:25) because acid treated filler had more surface area than non-acid treated filler. For example, before treated by acid CBe had surface area about 65.6 m^2/g and after treated by acid CBe had surface area about 98.85 m^2/g. Then filler had more surface area to contact with PP matrix it was better dispersability than the lower surface area (non-acid treated). From the morphology of composite, Particles of fillers in Figure 4.18 (b.) contact one another than in

Figure 4.18 (a.). Contact between filler is formed a continuous path for electrons to travel through. It was affect to the better movement of electrical current that cause higher electrical conductivity. However, it was the same results to previous work of Saranee [31]. She got higher electrical conductivity of composite that contain of acid treated filler. [31]

The effect of two types of conductive fillers had a greater result of electrical conductivity than one type of filler. The effect of acid – treated conductive fillers had a greater result of electrical conductivity than non-acid treated filler. The optimum ratio of part 1 and 2 was PP:ATAB:ATCBe (50:25:25) that had the highest electrical conductivity by in-plane (42.07 S/cm) and electrical conductivity by through-plane (39.12 S/cm) but still lower than the commercial requirement. The water absorption, density and shore hardness were passed the commercial requirement but the electrical conductivity and flexural strength were not passed the commercial requirement. And then, the optimum ratio (PP:ATAB:ATCBe in ratio of 50:25:25) was mixed with CNTs(Timesnano) and CNTs(N7000).

4.3 Study effect of mixture of carbon nanotubes

The optimum of part 1 and 2 (PP:ATAB:ATCBe in ratio of 50:25:25) was mixed with CNTs(Timesnano) and CNT(N7000)

4.3.1 Electrical properties

4.3.1.1 Electrical conductivity by in-plane technique

The electrical conductivity by in-plane technique of Polypropylene matrix and acid-treated conductive fillers (PP:ATAB:ATCBe) mixed with CNTs (Timesnano) and CNT (N7000) were shown in Figure 4.19

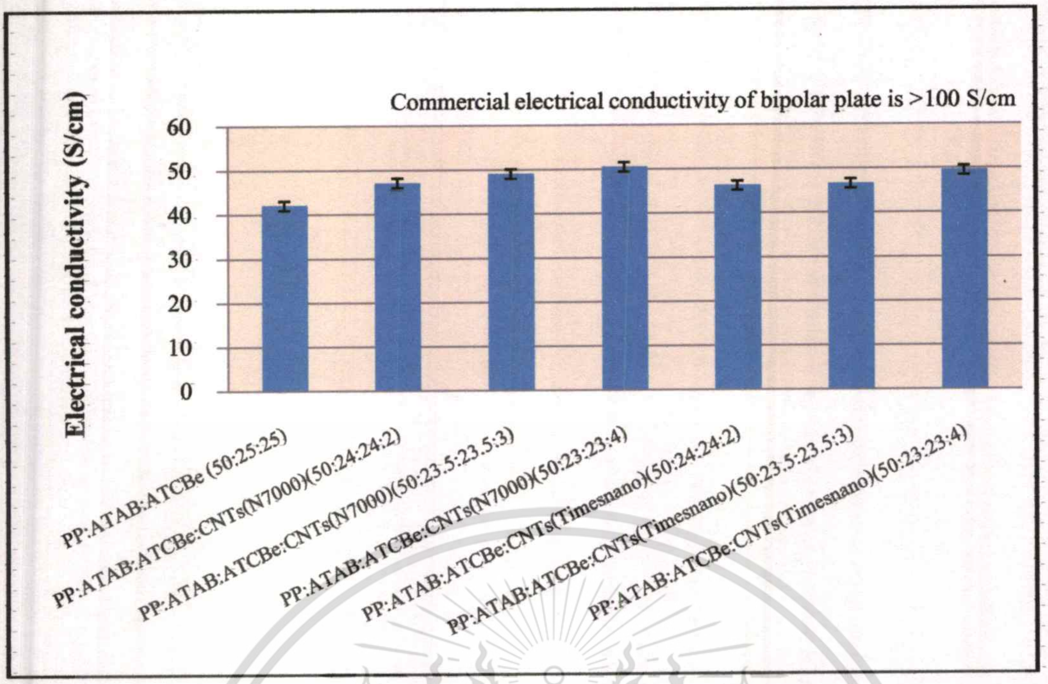


Figure 4.19 Electrical conductivity of Polypropylene matrix and conductive fillers (ATAB:ATCBe) with CNTs (Timesnano) and CNT (N7000)

Figure 4.19 shows electrical conductivity of Polypropylene matrix and conductive fillers (ATAB:ATCBe) with CNTs (Timesnano) and CNT (N7000) was in range of 46.23 – 50.51 S/cm. (Appendix D-1) The electrical conductivity of PP:ATAB:ATCBe was increased when it was mixed with CNTs. The CNT (N7000) filler provided higher electrical conductivity than CNTs (Timesnano) because their smaller size and higher electrical conductivity of 125 S/cm, compared to CNTs (Timesnano) had electrical conductivity of 100 S/cm. The lowest electrical conductivity was 46.23 S/cm (PP:ATAB:ATCBe:CNT(Timesnano);50:23:23:2). The highest electrical conductivity of the composite was 50.51 S/cm (PP:ATAB:ATCBe:CNT(N7000);50:23:23:4). There were also the effects of penetration ability of additives into the PP matrix, and the compatibility of fillers, CNT (N7000). Polymer composites doped with CNTs exhibit percolation behavior which presence of interconnected of carbon nanotube network results to increase their electricalconductivity more than polymer composite that without CNTs. However, at adding of 2% of CNTs was lower electrical conductivity than adding of 3% and 4% of CNTs because of the low number of contacts between nanotubes. To increase electrical conductivities, higher carbon nanotubes concentration may have more area contacts to produce higher conduction.

4.3.1.2 Electrical conductivity by through-plane technique

Electrical conductivity by through-plane technique of Polypropylene matrix and conductive fillers (ATAB:ATCBe) with CNTs (Timesnano) and CNT (N7000) were shown in Figure 4.20

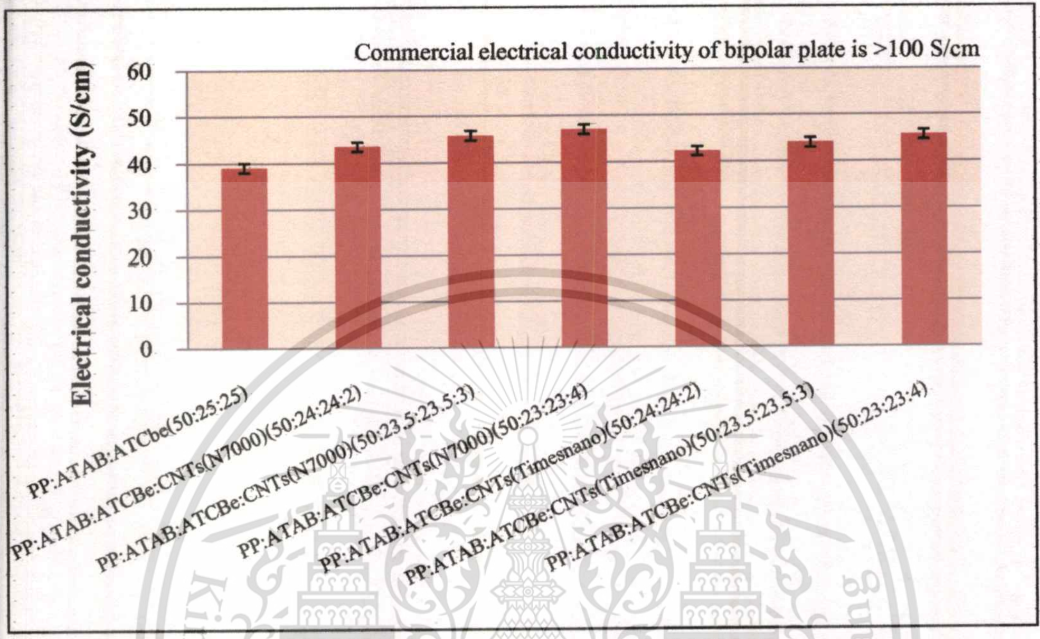


Figure 4.20 Electrical conductivity by through-plane technique of Polypropylene matrix and conductive fillers (ATAB:ATCBe) with CNTs (Timesnano) and CNT (N7000)

Figure 4.20 shows electrical conductivity by through-plane technique of Polypropylene matrix and conductive fillers (ATAB:ATCBe) with CNTs (Timesnano) and CNT (N7000) was in range of 42.38 – 47.12 S/cm. (Appendix D-2) The CNT (N7000) filler provided greater electrical conductivity than CNTs (Timesnano) because CNT (N7000) had smaller size than CNTs (Timesnano). The lowest electrical conductivity was 42.38 S/cm (PP:ATAB:ATCBe:CNTs(Timesnano) in ratio of 50:24:24:2) since the lowest amount of CNTs in the mixture of PP:ATAB:ATCBe. The highest electrical conductivity of the composite was PP: ATAB:ATCBe:CNT(N7000)(50:23:23:4) about 47.12 S/cm because of the highest amount of CNT(N7000) in the mixture of PP:ATAB:ATCBe. The tube of carbon nanotubes was connecting together and link with particle of acid-treated fillers that produce more path way to create the electrical current than non-carbon nanotube composite. However, adding 4% of CNTs was highest electrical conductivity because higher carbon nanotubes concentration may have more area contact between carbon nanotubes and acid treated conductive fillers (ATAB and ATCBe) to produce higher conduction. The through-plane electrical conductivity of the composite was lower than the in-plane electrical conductivity due to the particle of fillers are randomly distributed within the matrix and align in parallel pressure direction of the composite.

4.3.1.3 Resistivity by Through-plane technique

Interfacial Contact Resistance (ICR by Through-plane technique of Polypropylene matrix and conductives fillers (ATAB:ATCBe) with CNTs (Timesnano) and CNT (N7000) were shown in Figure 4.21.

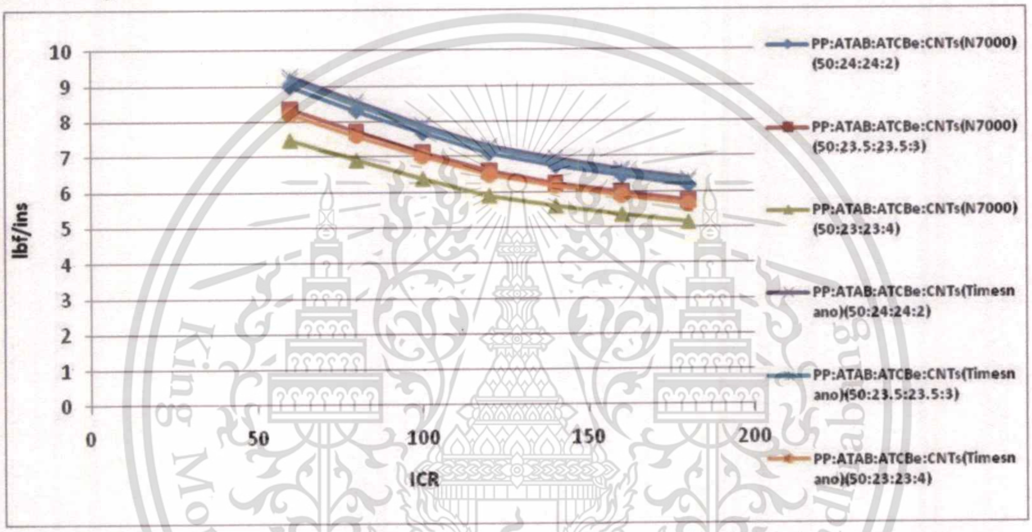


Figure 4.21 Interfacial contact resistance by Through-plane technique of Polypropylene matrix and conductive fillers (ATAB:ATCBe) with CNTs (Timesnano) and CNT (N7000)

Figure 4.21 shows the interfacial contact resistance (ICR) of PP that having different ratios of conducting fillers. The interfacial contact resistance (ICR) was decrease when they increase moment torque to the sample. The lower Interfacial, the higher electrical conductivity was. The lowest interfacial contact resistance dominated in PP: ATAB:ATCBe:CNT(N7000)(50:23:23:4) because of the smaller size and higher electrical conductivity of CNT(N7000). The structure of carbon nanotubes was like a long tube to connects together and link with particle of acid-treated fillers to produce more path way to create the electrical current than polymer composite that not contain of CNTs. However, adding 4% of

CNTs dominated the highest electrical conductivity because higher carbon nanotubes concentration may have more area contact between carbon nanotubes and acid treated conductive fillers (ATAB and ATCBe) to produce higher conduction. The through-plane electrical conductivity of the composite was lower than the in-plane electrical conductivity due to the particle of fillers are randomly distributed within the matrix and align in parallel pressure direction of the composite. (Appendix D-3)

4.3.2 Physical properties

4.3.2.1 Density

Volume and weight of Polypropylene matrix and conductives fillers (ATAB:ATCBe) with CNTs (Timesnano) and CNT (N7000) were measured by vernier caliper and then were calculated followed by equation (3.5). The results were shown in Figure 4.22

Density of Polypropylene matrix and conductive fillers (ATAB:ATCBe) with CNTs (Timesnano) and CNT (N7000) were shown in Figure 4.22

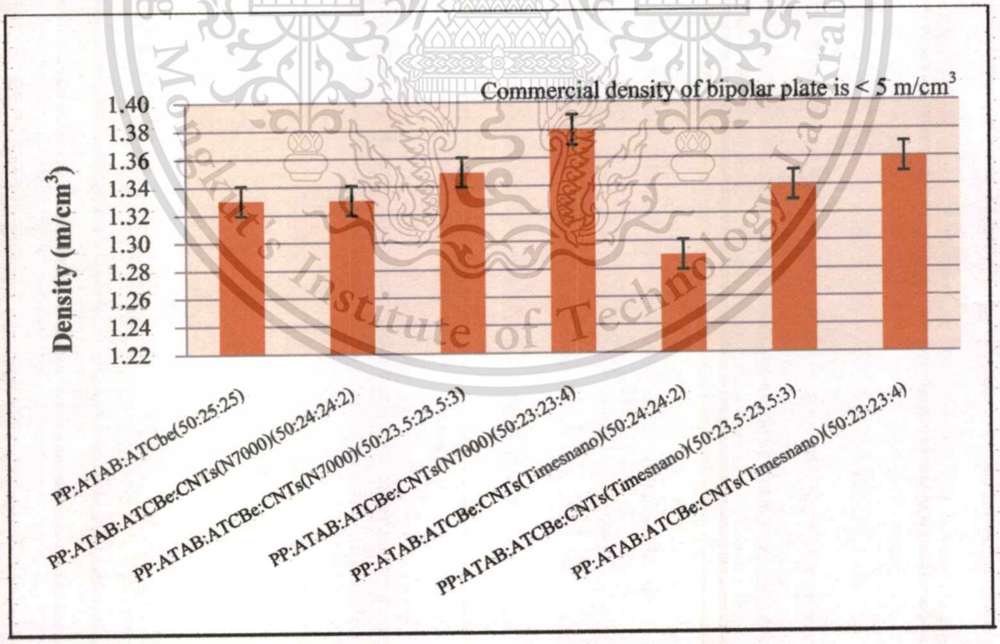


Figure 4.22 Density of Polypropylene matrix and conductives fillers (ATAB:ATCBe) in ratio of 50:23:23 with 4% of CNTs (Timesnano) and CNT (N7000).

The density of Polypropylene matrix and conductive fillers (ATAB:ATCBe) with CNTs (Timesnano) and CNT (N7000) (1.29-1.38 g/cm^3) was higher than the density of PP itself (0.90 g/cm^3) (Appendix D-4) and nearly the same to no adding carbon nanotubes.(Figure 4.22) The composites that contain of CNT (N7000) has the same range of density to CNTs (Timesnano). The trend of sample that contains CNT (N7000) was gradually increased from 1.33 g/cm^3 to 1.38 g/cm^3 as a percent of CNTs (N7000) were increased. The trend of sample that contains CNTs (Timesnano) was also increased from 1.29 g/cm^3 to 1.36 g/cm^3 when increased percent of CNTs (Timesnano). The density of PP:ATAB:ATCBe:CNTs(N7000) in ratio of 50:23:23:4 indicated the highest density of 1.38 g/cm^3. That was a little bit higher than PP:ATAB:ATCBe:CNTs (Timesnano) in ratio of 50:24:24:2 about 1.29 g/cm^3. Lower density is a key component, as a bipolar plate needs to be light and thin in order to reduce the overall cost and size of bipolar plate. The results indicated the low density that implied the good performance of bipolar plate in commercial. The density was between 1-2 g/cm3 was a common range of a good density. [40] The density of polymer composite that contain of CNTs was the same range to polymer composite that no contain CNTs because it was the amount of CNTs that adding to the polymer composite not much enough to change the density of composite. This was according to the penetration ability effect between fillers and matrix.

4.3.2.2 Water absorbtion of sample with ASTM D 570

The samples from compression molding were dried in an oven for removing moisture content for 24 hour at 80°C. After that, sample was marked as dry weight. Dry sample was put into water for 2 hours and marked as wet weight. Calculated water absorption followed by equation 3.6. The results were shown in Figure 4.23

Water absorbtion of Polypropylene matrix and conductive fillers (ATAB:ATCBe) with CNTs (Timesnano) and CNT (N7000) were shown in figure 4.23

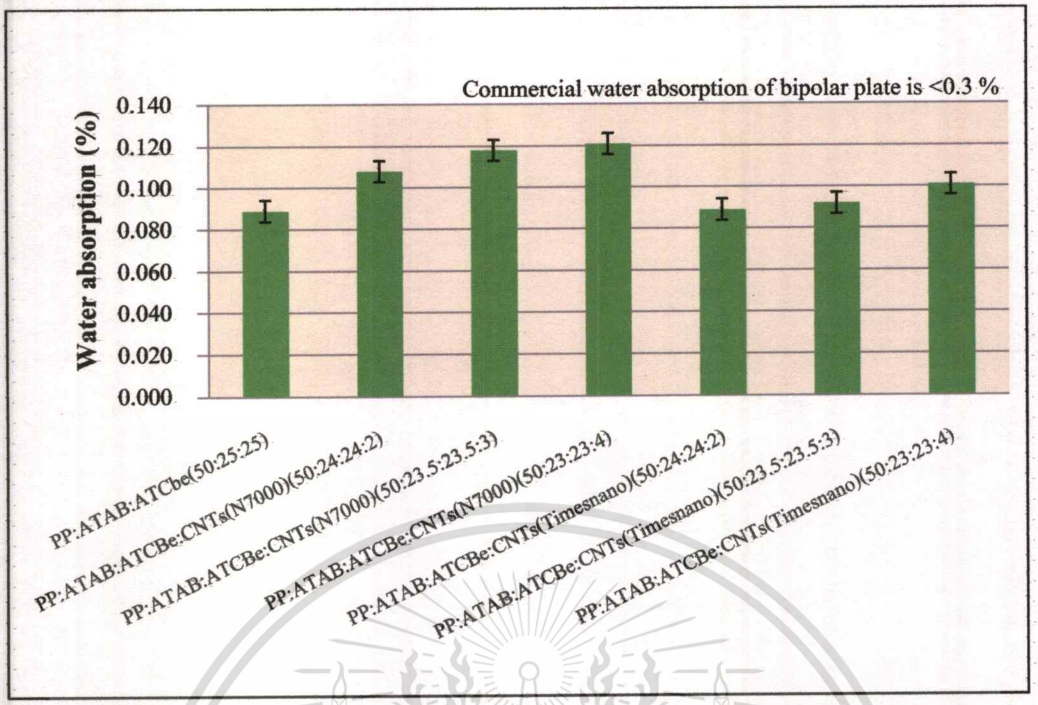


Figure 4.23 Water absorption of Polypropylene matrix and conductive fillers (ATAB:ATCBe) with CNTs (Timesnano) and CNT (N7000)

The tendency of the water absorption of PP:ATAB:ATCBe with 2-4 % of fillers (CNTs (Timesnano) and CNT (N7000) had the same trend but water absorption of CNT(N7000) was a little bit greater than CNTs (Timesnano) for the reason that CNT(N7000) had penetrated ability of PP:ATAB:ATCBe more than CNTs (Timesnano) (Appendix D-5). The lowest water absorption was PP: ATAB: ATCBe with 2% CNTs (Timesnano) was 0.089%. The highest water absorption was 0.108% PP: ATAB: ATCBe with 2% CNTs (N7000). The effect of CNT (N7000) was more dominated than CNTs (Timesnano). Actually, the increasing amount of mixture of CNTs had no significant effect to water absorption. In addition, the water absorption of this experiment could be applied in PEM fuel cell because PEM fuel cell requires less water absorption than 0.3 %.

4.3.3 Mechanical Properties

4.3.3.1 Hardness test

We prepared samples from compression molding. Shore D (HRD) was used to identify for Polypropylene matrix and conductives fillers (ATAB:ATCBe) with CNTs (Timesnano) and CNT (N7000) followed by ASTM D2240. It was shown in Figure 4.24.

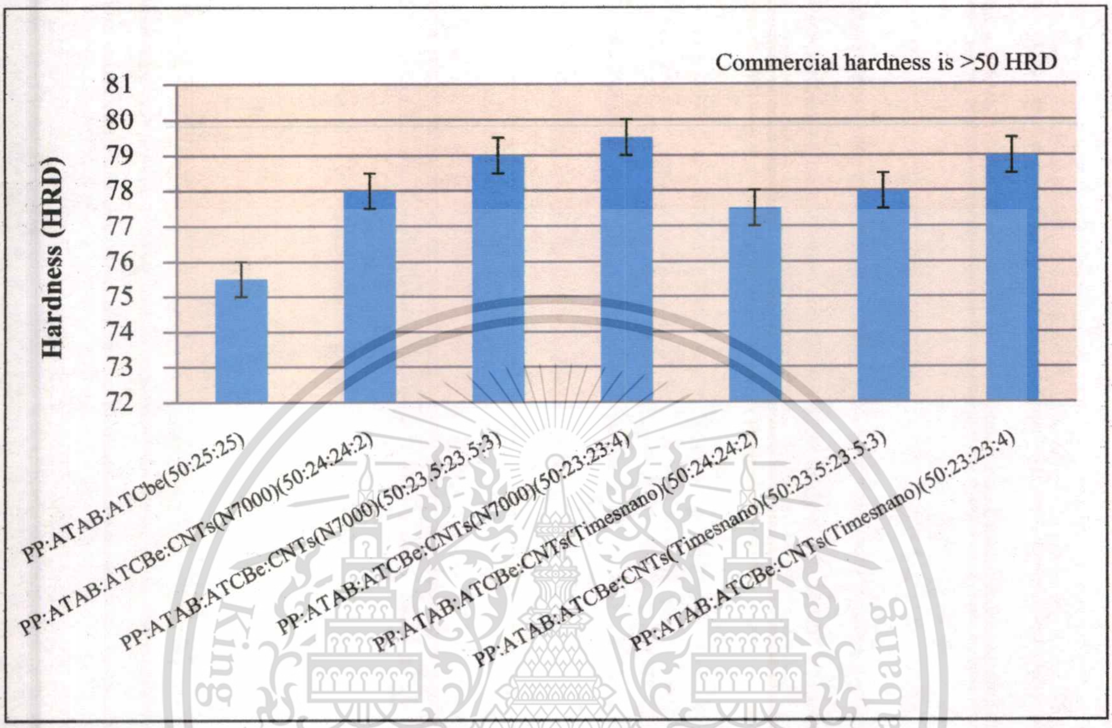


Figure 4.24 Hardness of Polypropylene matrix and conductives fillers (ATAB:ATCBe) with CNTs (Timesnano) and CNT (N7000)

The hardness of PP:ATAB:ATCBe with 2-4 % of fillers CNTs (Timesnano) and CNT (N7000) was 77.5 - 79.5 HRD. (Appendix D-6) The hardness of PP did not much change when fillers (CNTs (Timesnano) and CNT (N7000) were added. The highest hardness 79.5 HRD was dominated by PP:ATAB:ATCBe:CNTs(N7000) in ratio of 50:23:23:4. The lowest hardness 77.5 HRD was revealed by PP:ATAB:ATCBe:CNTs(Timesnano) in ratio of 50:24:24:2. This was due to penetration ability of acetylene black seated into the matrix PP. However, there are no significant changes in hardness of PP and conductive fillers (CNTs (Timesnano) and CNT (N7000). Hardness of polypropylene matrix and conductive fillers (ATAB:ATCBe) with CNTs (Timesnano) and CNTs (N7000) was more than no adding carbon nanotubes because hardness of materials was depend on the type of forces that combined between atoms in the material. The tube structure of carbon nanotube was created the strong linkages at the interface between phase's CNTs and PP matrix result to increase the hardness. Therefore, the hardness of composites was passed through the commercial requirement (> 50 HRD).

4.3.3.2 Flexural strength

The flexural strength of composite plate was measured by three point bending test using a universal testing machine followed by ASTM D790-03. Rectangular polypropylene composites of height 10 mm. width 3 mm. and length 120 mm. in dimensions were placed on two supports with span length 50 mm and rate about 10 mm./minute. A load at the midpoint of the samples was initiated in the way that is presented in Figure 4.25 Maximum stress and strains were calculated on the incremental load applied for the evaluation of bending strength.

The flexural strength of the Polypropylene matrix and conductive fillers (ATAB:ATCBe) in ratio of 50:25:25 with CNTs (Timesnano) and CNT (N7000)

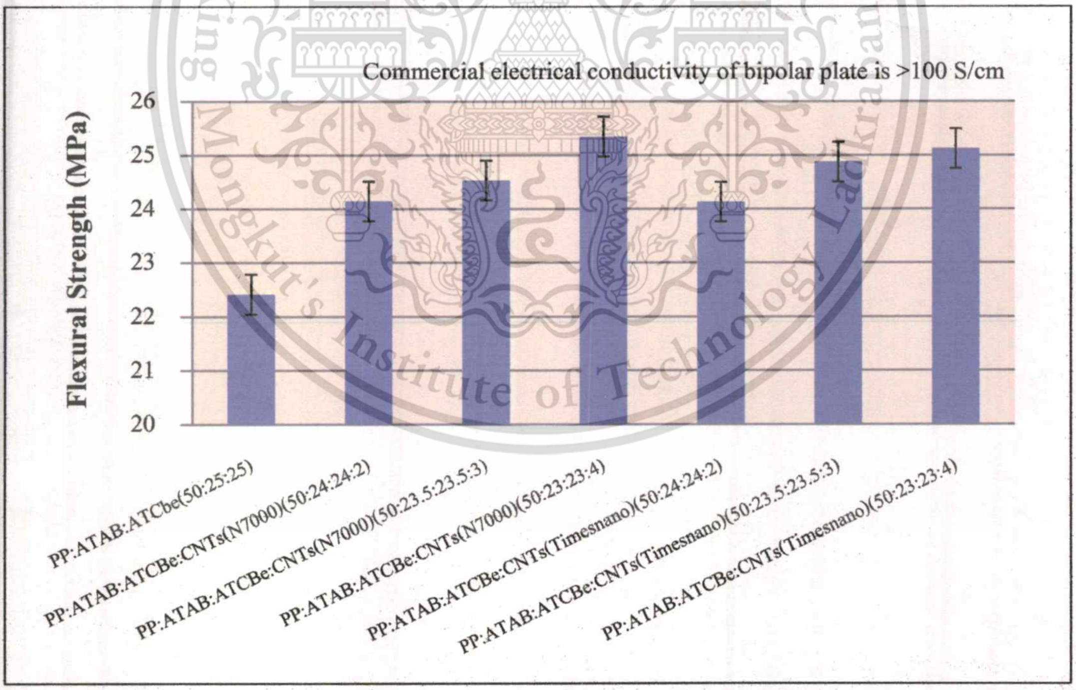
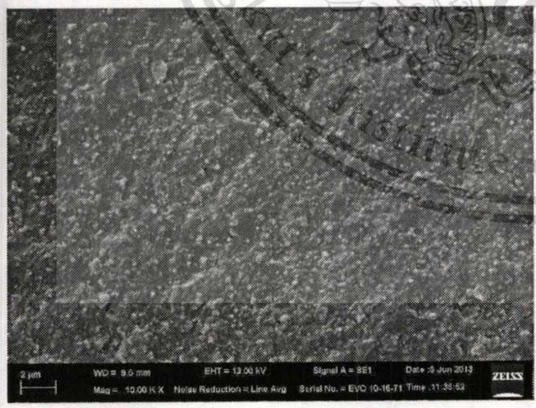


Figure 4.25 Flexural strength of the Polypropylene matrix and conductives fillers (ATAB:ATCBe) in ratio of 50:25:25 with CNTs(Timesnano) and CNT(N7000)

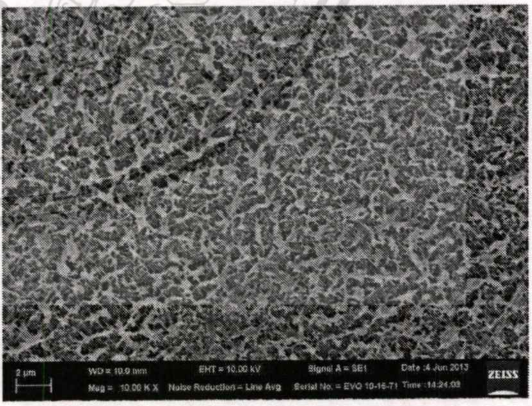
Flexural strength varied with different fillers was shown in Figure 4.25. The flexural strength increased with adding more amounts of CNTs (Timesnano) and CNT (N7000). The measured flexural strength for the polymer composite of bipolar plates was in range of 24.13 – 25.35 MPa. (Appendix D-7) The results were indicated that the addition of CNT (N7000) was greater flexural strength than of CNTs (Timesnano), the addition of PP:ATAB:ATCBe with 4 % of fillers (CNT(N7000) was revealed the greatest effect of 25.35 MPa. The lowest flexural strength (24.13 MPa) was PP:ATAB:ATCBe:CNTs(Timesnano) in ratio of 50:24:24:2). Therefore, the flexural strength of composites was not passed through the commercial requirement (> 59 MPa). Flexural strength of no adding CNTs composite (PP:ATAB:ATAB) was found to be 22.41 MPa. The polymer composite with CNTs shows greater flexural strength than no adding CNTs about 24.13 – 25.35 MPa. This was due to the average ratio and high strength of CNTs, their dispersion, and the good interfacial bonding between CNTs and the polymer matrix contributed to the increase in the flexural properties of the composite produced.

4.3.4 Morphology by Scanning Electron Microscope (SEM)

A scanning Electron Microscope (SEM) technique was an effective way to view the microstructure of composite for explanation dispersability of Polypropylene matrix and conductive fillers (ATAB:ATCBe) with CNTs (Timesnano) and CNT (N7000). The results were shown in Figure 4.26.

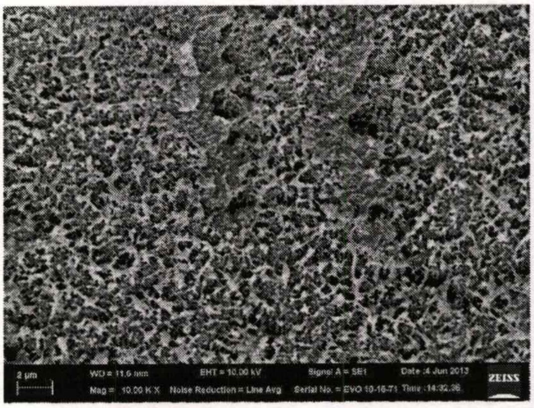


a.)PP:ATAB:ATCBe(50:25:25)



b.)PP:ATAB:ATCBe:CNTs(N7000)

(50:23:23:4)



c.)PP:ATAB:ATCBe:CNTs(Timesnano) (50:23:23:4)

Figure 4.26 Morphology of Polypropylene matrix and conductives fillers (ATAB:ATCBe) with CNTs (Timesnano) and CNT (N7000) with magnification 10,000 times. a.)PP:ATAB:ATCBe (50:25:25) b.)PP:ATAB:ATCBe:CNTs(N7000) (50:23:23:4) c.)PP:ATAB:ATCBe:CNTs(Timesnano) (50:23:23:4)

Figure 4.26 shows the SEM images of the surfaces of PP:ATAB:ATCBe with adding of CNTs (N7000) and CNTs (Timesnano). Polypropylene composite in Figure 4.26(b and c) was indicated higher electrical conductivity when compared to polypropylene composite in Figure 4.26 (a.). It was due to the nano – sized CNTs being the filler that have a tube shape and being able to fill the pores and connect between ATAB and ATCBe in PP matrix. Thus, the tube of carbon nanotubes was connecting together and contact with particle of acid-treated fillers that produce more path way to create the continuous path for electrical current than non-carbon nanotubes composite. The optimum dispersability was polypropylene composite in Figure 4.26 (b.) that adding of CNTs (N7000) because CNTs (N7000) had smaller diameter of tube and more purity than CNTs (Timenano) in Figure 4.26 (c.). Therefore, it was contact with ATAB, ATCBe and PP matrix better than a bigger diameter (CNTs (Timesnano)). From the morphology of composite in Figure .26 (b,c), the good interfacial bonding between high strength CNTs, acid-treated fillers and the polymer matrix contributed to increase in the flexural properties of composite. So, Figure 4.26 (b,c) was higher flexural strength than in Figure 4.26 (a.). Therefore, the strong linkages at the interface between phase's CNTs and PP matrix result to increase the hardness of composite that contain of CNTs (Figure 4.26 (a.).

4.4 Comparison of properties between commercial and polypropylene composite bipolar plate

Table 4.1 Comparison of properties between commercial and polypropylene composite bipolar plate.

	Commercial	Polypropylene composite bipolar plate (PP:ATAB:ATCBe:CNT(N7000) in ratio of 50:23:23:4)
In-plane electrical conductivity (S/cm)	> 100	50.51
Flexural Strength (Mpa)	> 59	25.35
Water absorption (%)	< 0.3%	0.12
Density (g/cm^3)	< 5	1.38
Shore hardness (HRD)	> 50	79.5

By comparison to the commercial bipolar plates, water absorption, density and shore hardness were accepted in commercial use (Table 4.1). However, to meet the commercial requirement, in-plane electrical conductivity and flexural strength need to be improved.

Chapter 5

Conclusion and recommendation

5.1 Conclusion

We prepared bipolar plate from composites polymer. The matrix was PP with various ratios of conductive fillers (CBn,AB,GT,CBe), acid-treated conductive fillers (ATCBn,ATAB,ATGT,ATCBe) and CNTs (Timesnano,N7000). The two-roll mill was used for blending. After that, composites were ground with grinder machine. Finally, they were molded with compression molding. The conclusion of polymer composite can be separated into three parts. First part was studied the effect of conductive fillers (CBn,AB,GT,CBe) to Polypropylene matrix. Second part was studied the effect of acid-treated conductive fillers (ATCBn, ATAB,ATGT,ATCBe) to Polypropylene matrix. Third part was studied the effect of CNTs (Timesnano) and CNT (N7000) to the optimum of Polymer composite (PP:ATAB:ATCBe) from part 1 and 2.

1. The effect of two types of non-acid treated conductive fillers (CBn, AB, GT, CBe) have a greater result than one type of non-acid treated fillers (CBn, AB, GT, CBe). The effect of non-acid treated conductive fillers (CBn, AB, GT, CBe) to polypropylene matrix was found that the optimum electrical conductivity by in-plane technique (PP:CBe:aB(50:25:25)) was 40.93 S/cm. and the electrical conductivity by through-plane technique (PP:CBe:AB(50:25:25)) was 37.47 S/cm. The results of density (1.08 – 1.40 g/cm^3), water absorption (0.067 – 1.114 %.) and hardness (72 – 78.5 HRD) were in a good range. The result of flexural strength was not good. The flexural strengths were in range of 20.09 – 29.25 MPa.

2. The effect of acid-treated conductive fillers (ATCBn, ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25 to Polypropylene matrix was found that the optimum electrical conductivity was more than polymer composite with non-acid treated filler. The optimum electrical conductivity was two-types of acid-treated conductive fillers PP:ATCBe:ATAB (50:25:25) that had electrical conductivity by in-plane technique about 42.07 S/cm and the electrical conductivity by through-plane technique about 39.12 S/cm. The result of density (1.10 – 1.47 g/cm^3), water

absorption (0.063 – 0.105%) and hardness (7.15 – 77 HRD) was in a good range. The flexural strength was still low in range of 19.05 – 26.44 MPa.

3. The effect of CNT's (Timesnano) and CNT(N7000) was found that the optimum electrical conductivity was more than polymer composite with non-CNTs. The optimum electrical conductivity by in-plane technique was PP:ATCBe:ATAB:CNTs(N7000) (50:23:23:4) (50.51 S/cm) that had the electrical conductivity by through-plane technique about 47.12 S/cm. result of density (1.29.g/cm^3), water absorption (0.089 – 0.121%) and hardness (75.5 – 79.5 HRD) was in good range. The flexural strength was in range of 22.41- 25.35 MPa, it was more than non-CNTs but still low.

By comparison of properties between commercial bipolar plate and polypropylene composite bipolar plate, water absorption, density and hardness were accepted in commercial use. But electrical conductivity and flexural strength still lower than commercial requirement.

5.2 Recommendation

1. Internal mixing is recommended for prevention of lost material during process because two-roll mill could not control dispersability and temperature.

2. Composites used in PEMs fuel cell should be strong; carbon fiber is the good chance to improve mechanical properties.

3. The electrical conductivity is depend on matrix and ratio of additives, the selection of new matrix should be investigated for enhance electrical conductivity.

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Appendix A

A-1 Electrical conductivity by in-plane technique

Information of experimental

Polypropylene matrix and conductives fillers (ATAB:ATCBe) with CNT (N7000) in ratio of 50:23:23:4 (Table A-1)

Length between electrodes (L) = 1 mm.

Electric current (I) = 0.012 A

Cross-sectional area (A) = 0.3

Table A-1 Electrical potential (V) of Polypropylene matrix and conductives fillers (ATAB:ATCBe) with CNT (N7000) in ratio of 50:23:23:4

Electrical potential (V)				
Trial 1	Trial 2	Trial 3	Trial 4	Trial 5
0.0000788	0.0000795	0.0000793	0.0000791	0.0000793

Average of electrical potential (Vave) = $\frac{(0.0000788+0.0000795+0.0000793+0.0000791+0.0000793)}{5}$

$= 0.0000792$ V

Calculate of electrical resistance (R) from equation (3.1)

$$R = \frac{V}{I} \tag{3.1}$$

$$R = \frac{0.0000792}{0.012}$$

$$= 0.0066\ \Omega$$

Calculate of electrical resistivity (ρ) of from equation (3.2)

$$\rho = \frac{R*A}{L} \tag{3.2}$$

$$= \frac{0.0066*0.3}{0.1}$$

$$= 0.0198$$

Calculate of conductivity (σ) of sample 23from equation (3.3)

$$\sigma = \frac{1}{\rho} \tag{3.3}$$

$$\sigma = \frac{1}{0.0198}$$

$$= 50.505 \text{ S.cm}^{-1}$$

From graph $R_2 = 0.0064$

$$ICR = \frac{R_2 - R_1}{2} = \frac{0.0064 - 0.003}{2} = 0.0017\,\Omega$$

Calculate of electrical resistivity from equation (3.2)

Cross-sectional area (A) = 3.5 cm^2

Thickness of sample (L) = 0.3 cm^2

$$\rho = \frac{RxA}{L} = \frac{0.0017x3.5}{0.3} = 0.01983\,\Omega$$

Calculate electrical conductivity of sample from equation (3.3)

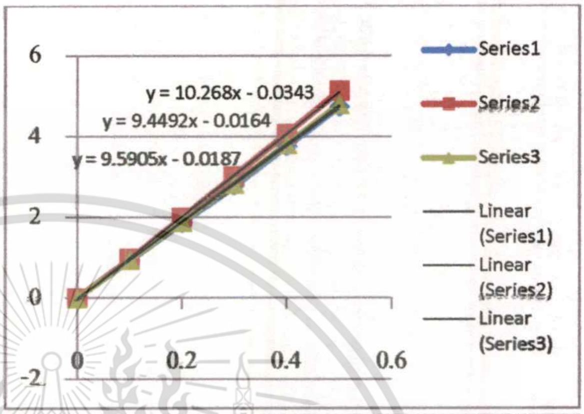
$$\sigma = \frac{1}{\rho} = \frac{1}{\rho} = 50.42\ Scm^{-1}$$

A-3 Resistivity by through-plane technique

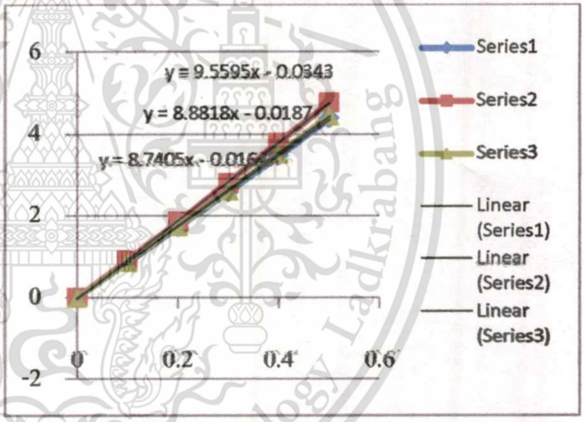
Interfacial contact resistance (ICR) of (PP:ATAB:ATCBe:CNTs(N7000) in ratio of 50:23:23:4)

Plot a graph between Potential and electric current, and then, calculate of Resistivity by equation V=IR with different load.

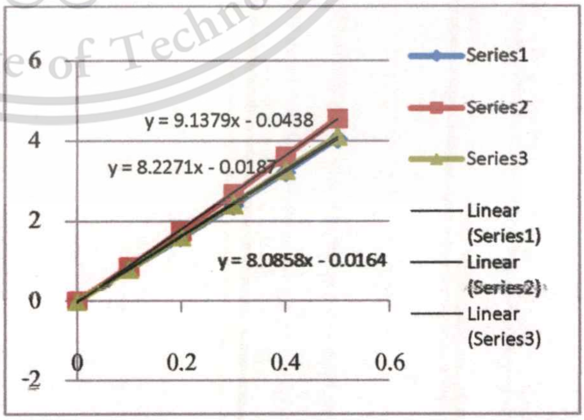
60 lbf/ins	Sample1	Sample2	Sample3
I(A)	V(mV)	V(mV)	V(mV)
0	0	0	0
0.1	0.930905	0.978905	0.941905
0.2	1.86181	1.99381	1.88621
0.3	2.792715	3.026715	2.831415
0.4	3.75822	4.07162	3.81202
0.5	4.731875	5.125525	4.802239
R	9.4492	10.268	9.5905



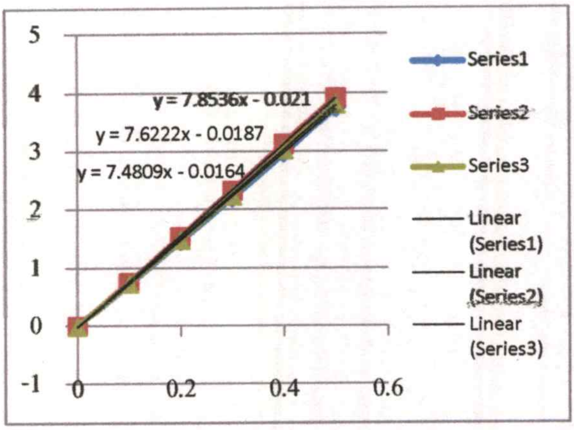
80 lbf/ins	Sample1	Sample2	Sample3
I(A)	V(mV)	V(mV)	V(mV)
0	0	0	0
0.1	0.860038	0.908038	0.871038
0.2	1.720076	1.852076	1.744476
0.3	2.580113	2.814113	2.618813
0.4	3.474751	3.788151	3.528551
0.5	4.377539	4.771189	4.447903
R	8.7405	9.5595	8.8818



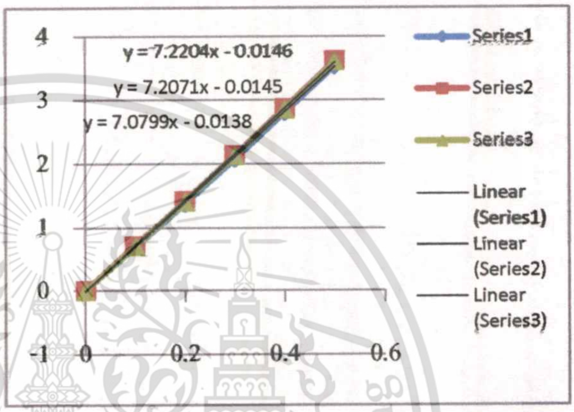
100 lbf/ins	Sample1	Sample2	Sample3
I(A)	V(mV)	V(mV)	V(mV)
0	0	0	0
0.1	0.794566	0.852566	0.805566
0.2	1.589131	1.751131	1.613531
0.3	2.383697	2.672697	2.422397
0.4	3.212862	3.609596	3.266662
0.5	4.050178	4.557995	4.120542
R	8.0858	9.1379	8.2271



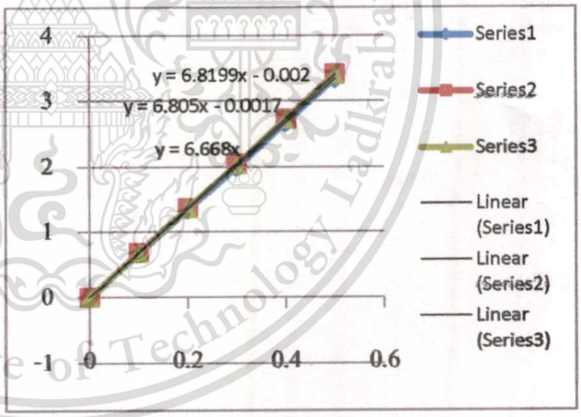
120 lbf/ins	Sample1	Sample2	Sample3
I(A)	V(mV)	V(mV)	V(mV)
0	0	0	0
0.1	0.734078	0.756078	0.745078
0.2	1.468155	1.534155	1.492555
0.3	2.202233	2.323233	2.240933
0.4	2.970911	3.119644	3.024711
0.5	3.747738	3.921555	3.818102
R	7.4809	7.8536	7.6222



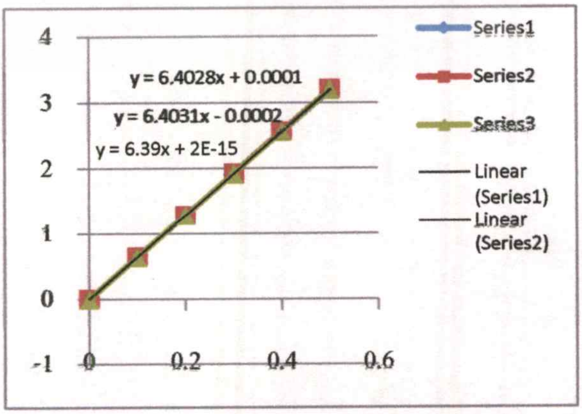
140 lbf/ins	Sample1	Sample2	Sample3
I(A)	V(mV)	V(mV)	V(mV)
0	0	0	0
0.1	0.696402	0.708402	0.707402
0.2	1.392805	1.419605	1.417205
0.3	2.089207	2.132207	2.127907
0.4	2.811009	2.865242	2.859479
0.5	3.547861	3.617673	3.611563
R	7.0799	7.224	7.2071



160 lbf/ins	Sample1	Sample2	Sample3
I(A)	V(mV)	V(mV)	V(mV)
0	0	0	0
0.1	0.666797	0.678797	0.677797
0.2	1.333593	1.360393	1.357993
0.3	2.00039	2.04339	2.03909
0.4	2.667186	2.726819	2.721056
0.5	3.333983	3.408524	3.40132
R	6.668	6.8199	6.805



180 lbf/ins	Sample1	Sample2	Sample3
I(A)	V(mV)	V(mV)	V(mV)
0	0	0	0
0.1	0.639	0.641	0.64
0.2	1.278	1.28	1.28
0.3	1.917	1.92095	1.92081
0.4	2.556	2.561267	2.56108
0.5	3.195	3.201583	3.20135
R	6.39	6.4028	6.4031



Interfacial contact resistance was depend on R and A; So, Interfacial contact resistance was equal to RxA.

Interfacial contact resistance of machine = R_1

Interfacial contact resistance of sample =R_2

Interfacial contact resistance (ICR) = $(R_2-R_1)/2$

	Electrical resistance (R)			Average of resistance	Standard diviation		R2	R1	ICR of sample
Lbf/ins	Sample 1	Sample 2	Sample 3			Area			
60	9.4492	10.268	9.5905	9.77	0.4377	3	29.3077	14.361	7.47
80	8.7405	9.5595	8.8818	9.06	0.5791	3	27.1818	13.3728	6.9
100	8.0858	9.1379	8.2271	8.48	0.7439	3	25.4508	12.693	6.38
120	7.4809	7.8536	7.6222	7.65	0.2635	3	22.3567	11.1702	5.89
140	7.0799	7.224	7.2071	7.17	0.1019	3	21.511	10.3256	5.59
160	6.668	6.8199	6.805	6.76	0.1074	3	20.2929	9.5865	5.35
180	6.39	6.4028	6.4031	6.40	0.0091	3	19.1959	8.9359	5.13

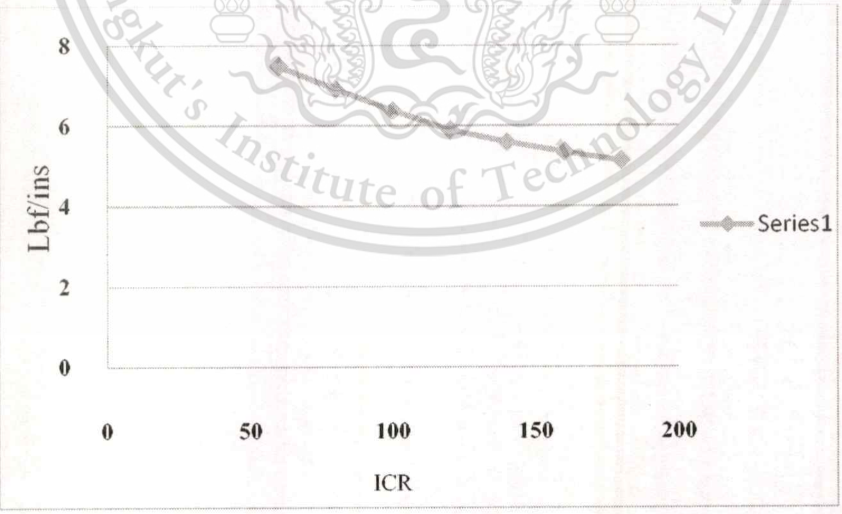


Figure A1 Interfacial contact resistance (ICR) of PP:ATAB:ATCBe:CNTs(N7000) in ratio of 50:23:23:4

A-4 Density

Density of of PP:ATAB:ATCBe:CNTs(N7000) in ratio of 50:23:23:4 was calculated by equation 3.5

$$\text{Density (D)} = \frac{\text{Mass (M)}}{\text{Volume (V)}} \qquad (3.5)$$

$$\text{Mass (M)} = 0.828 \text{ g}$$

$$\text{Volume (V)} = 0.6 \text{ cm}^3$$

$$\text{Density (D)} = \frac{0.828}{0.6} \text{ g/cm}^3$$

$$= 1.38 \text{ g/cm}^3$$

Appendix B

Study effect of conductive fillers (CBn,AB,GT,CBe) in ratio of 50:50 and 50:25:25 to Polypropylene matrix (Table B-1 – B-7)

Table B-1 Electrical conductivity of conductive fillers (CBn,AB,GT,CBe) in ratio of 50:50 and 50:25:25 to Polypropylene matrix by in-plane technique

Ratio of PP matrix and additives	Trial1 (S/cm)	Trial2 (S/cm)	Trial3 (S/cm)	Trial4 (S/cm)	Trial5 (S/cm)	Average (S/cm)	S.D.
PP:AB(50:50)	35.40	35.71	35.09	34.78	39.97	36.19	2.14
PP:CBn(50:50)	35.40	35.72	35.09	35.52	34.78	35.30	0.37
PP:GT(50:50)	31.25	31.01	31.75	30.89	29.56	30.89	0.81
PP:Cbe(50:50)	39.25	39.02	39.57	38.28	39.98	39.22	0.64
PP:AB:CBn(50:25:25)	37.21	37.38	36.97	37.23	35.26	36.81	0.88
PP:AB:GT(50:25:25)	36.27	39.62	33.59	35.86	36.02	36.27	2.16
PP:AB:Cbe(50:25:25)	40.94	41.09	40.91	40.49	41.23	40.93	0.28
PP:CBn:GT(50:25:25)	32.03	31.97	32.22	32.01	31.87	32.02	0.13
PP:CBn:Cbe(50:25:25)	34.48	33.33	34.34	34.13	34.52	34.16	0.49
PP:GT:Cbe(50:25:25)	33.03	33.28	33.32	33.14	33.18	33.19	0.12

Table B-2 Electrical conductivity by Through-plane technique of Polypropylene matrix and conductive fillers (CBn, AB,GT,CBe) in ratio of 50:50 and 50:25:25

Ratio of PP matrix and additives	Trial1 (S/cm)	Trial2 (S/cm)	Trial3 (S/cm)	Trial4 (S/cm)	Trial5 (S/cm)	Average (S/cm)	S.D.
PP:AB(50:50)	31.52	31.65	32.49	32.09	33.96	32.34	0.980
PP:CBn(50:50)	30.15	30.13	31.65	31.26	31.43	30.92	0.729
PP:GT(50:50)	24.26	23.57	24.70	23.44	25.48	24.29	0.838
PP:Cbe(50:50)	36.46	35.21	35.08	35.55	36.00	35.66	0.569
PP:AB:CBn(50:25:25)	33.01	33.25	32.25	33.15	33.49	33.03	0.467
PP:AB:GT(50:25:25)	32.15	32.05	33.15	33.57	30.29	32.24	1.267
PP:AB:Cbe(50:25:25)	37.25	36.49	37.45	36.49	39.67	37.47	1.306
PP:CBn:GT(50:25:25)	26.50	26.48	27.07	25.35	24.76	26.03	0.945
PP:CBn:Cbe(50:25:25)	28.97	27.35	27.65	29.64	31.29	28.98	1.599
PP:GT:Cbe(50:25:25)	28.64	28.05	27.31	27.34	27.91	27.85	0.554

Table B-3 Interfacial Contact Resistance (ICR by Through-plane technique of Polypropylene matrix and conductive fillers (CBn, AB,GT,CBe) in ratio of 50:50 and 50:25:25

Ratio of PP matrix and additives	ICR in different moment torque						
	60 lbf/ins	80 lbf/ins	100 lbf/ins	120 lbf/ins	140 lbf/ins	160 lbf/ins	180 lbf/ins
PP:AB(50:50)	13.64	12.60	11.64	10.75	10.20	9.77	9.36
PP:CBn(50:50)	14.14	13.06	12.06	11.15	10.58	10.13	9.71
PP:GT(50:50)	17.09	15.79	14.59	13.48	12.78	12.24	11.73
PP:Cbe(50:50)	12.08	11.16	10.31	9.53	9.04	8.66	8.30
PP:AB:CBn(50:25:25)	13.31	12.29	11.36	10.49	9.96	9.53	9.14
PP:AB:GT(50:25:25)	13.59	12.56	11.60	10.72	10.17	9.74	9.33
PP:AB:Cbe(50:25:25)	11.30	10.44	9.64	8.91	8.45	8.09	7.76
PP:CBn:GT(50:25:25)	16.26	15.02	13.88	12.82	12.16	11.65	11.16
PP:CBn:Cbe(50:25:25)	14.84	13.71	12.66	11.70	11.10	10.63	10.19
PP:GT:Cbe(50:25:25)	15.45	14.27	13.19	12.18	11.56	11.07	10.61

Table B-4 The density of Polypropylene and conductive fillers (CBn,AB,GT,CBe) in ratio of 50:50 and 50:25:25

Ratio of PP matrix and additives	Trial 1	Trial 2	Trial 3	Trial 4	Density	S.D
	Density (g/cm3)	Density (g/cm3)	Density (g/cm3)	Density (g/cm3)	average (g/cm3)	
PP:AB(50:50)	1.21	1.25	1.28	1.34	1.27	0.055
PP:CBn(50:50)	1.35	1.31	1.32	1.50	1.37	0.088
PP:GT(50:50)	1.1	1.05	1.04	1.13	1.08	0.042
PP:Cbe(50:50)	1.15	1.17	1.12	1.08	1.13	0.039
PP:AB:CBn(50:25:25)	1.38	1.39	1.42	1.41	1.40	0.018
PP:AB:GT(50:25:25)	1.19	1.23	1.21	1.25	1.22	0.026
PP:AB:Cbe(50:25:25)	1.16	1.2	1.19	1.17	1.18	0.018
PP:CBn:GT(50:25:25)	1.22	1.24	1.2	1.26	1.23	0.026
PP:CBn:Cbe(50:25:25)	1.19	1.17	1.16	1.20	1.18	0.018
PP:GT:Cbe(50:25:25)	1.14	1.16	1.17	1.05	1.13	0.055

Table B -5 Water absorption of Polypropylene and conductives fillers (CBn, AB,GT,CBe) in ratio of 50:50 and 50:25:25

Ratio of PP matrix and additives	Trial 1	Trial 2	Trial 3	Trial 4	Average	S.D
	Water absorption (%)	Water absorption (%)	Water absorption (%)	Water absorption (%)	Water absorption (%)	
PP:AB(50:50)	0.085	0.087	0.091	0.097	0.090	0.005
PP:CBn(50:50)	0.074	0.072	0.078	0.080	0.076	0.004
PP:GT(50:50)	0.092	0.089	0.091	0.100	0.093	0.005
PP:Cbe(50:50)	0.065	0.069	0.064	0.070	0.067	0.003
PP:AB:CBn(50:25:25)	0.104	0.101	0.1	0.103	0.102	0.002
PP:AB:GT(50:25:25)	0.115	0.112	0.114	0.115	0.114	0.001
PP:AB:Cbe(50:25:25)	0.085	0.091	0.087	0.093	0.089	0.004
PP:CBn:GT(50:25:25)	0.091	0.094	0.093	0.090	0.092	0.002
PP:CBn:Cbe(50:25:25)	0.081	0.085	0.086	0.088	0.085	0.003
PP:GT:Cbe(50:25:25)	0.086	0.089	0.091	0.090	0.089	0.002

Table B-6 Hardness of Polypropylene and conductives fillers (CBn,AB,GT,CBe) in ratio of 50:50 and 50:25:25

Ratio of PP matrix and additives	Trial1	Trial2	Trial3	Trial4	Trial5	Average HRD	S.D.
	HRD	HRD	HRD	HRD	HRD		
PP:AB(50:50)	77	76.5	76	77	76	76.5	0.500
PP:CBn(50:50)	74	74.5	74	75	75	74.5	0.500
PP:GT(50:50)	75	74.5	75	74	76.5	75	0.935
PP:Cbe(50:50)	71.5	72	72	71.5	73	72	0.612
PP:AB:CBn(50:25:25)	77.5	78	78	77	79.5	78	0.935
PP:AB:GT(50:25:25)	78	77.5	78	78.5	80.5	78.5	1.173
PP:AB:Cbe(50:25:25)	77	77.5	77	78	75.5	77	0.935
PP:CBn:GT(50:25:25)	77.5	77	77	78	78	77.5	0.500
PP:CBn:Cbe(50:25:25)	74.5	75	75	76	74.5	75	0.612
PP:GT:Cbe(50:25:25)	75.5	76	76	77	75.5	76	0.612

Table B-7 The flexural strength of the polypropylene with conductive fillers (CBn,AB,GT,CBe) in ratio of 50:50 and 50:25:25

Ratio of PP matrix and additives	Trial1	Trial2	Trial3	Trial4	Trial5	Average	S.D.
	Flexural Strength (Mpa)	Flexural Strength (Mpa)	Flexural Strength (Mpa)	Flexural Strength (Mpa)	Flexural Strength (Mpa)	Flexural Strength (Mpa)	
PP:AB(50:50)	24.53	25.65	24.35	24.26	24.47	24.65	0.565
PP:CBn(50:50)	20.22	19.47	20.34	20.45	20.58	20.21	0.439
PP:GT(50:50)	25.43	25.36	25.42	25.36	26.34	25.58	0.426
PP:Cbe(50:50)	20.09	20.35	20.49	19.65	19.86	20.09	0.343
PP:AB:CBn(50:25:25)	26.21	26.44	25.43	25.85	28.21	26.43	1.067
PP:AB:GT(50:25:25)	29.25	28.36	29.21	28.65	30.76	29.25	0.925
PP:AB:Cbe(50:25:25)	25.74	25.67	25.36	24.68	26.77	25.64	0.757
PP:CBn:GT(50:25:25)	27.22	27.47	26.31	27.12	27.50	27.12	0.482
PP:CBn:Cbe(50:25:25)	24.58	24.66	24.36	23.56	26.74	24.78	1.179
PP:GT:Cbe(50:25:25)	25.88	26.55	26.42	26.46	27.13	26.49	0.447

Appendix C

Study effect of conductive fillers (ATCBn,ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25 in Polypropylene matrix (Table C-1 – C-7)

Table C-1 Electrical conductivity of conductive fillers (ATCBn,ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25 to Polypropylene matrix by in-plane technique

Ratio of PP matrix and additives	Trial1	Trial2	Trial3	Trial4	Trial5	Average	S.D.
	(S/cm)	(S/cm)	(S/cm)	(S/cm)	(S/cm)	(S/cm)	
PP:ATAB(50:50)	37.74	37.77	37.49	37.42	37.54	37.59	0.16
PP:ATCBn(50:50)	35.37	35.09	35.37	35.52	35.50	35.37	0.17
PP:ATGT(50:50)	32.00	32.49	31.95	32.03	32.13	32.12	0.22
PP:ATCbe(50:50)	40.36	40.86	40.90	40.61	40.52	40.65	0.23
PP:ATAB:ATCBn(50:25:25)	38.80	39.41	38.95	38.95	39.00	39.02	0.23
PP:ATAB:ATGT(50:25:25)	37.21	37.11	37.24	37.00	36.84	37.08	0.16
PP:ATAB:ATCbe(50:25:25)	42.50	42.10	42.51	42.46	40.79	42.07	0.74
PP:ATCBn:ATGT(50:25:25)	34.16	34.13	33.87	33.73	34.02	33.98	0.18
PP:ATCBn:ATCbe(50:25:25)	36.80	36.70	36.80	37.07	37.13	36.90	0.19
PP:ATGT:ATCbe(50:25:25)	34.16	34.28	34.10	34.31	34.26	34.22	0.09

Table C-2 Electrical conductivity by Through-plane technique of Polypropylene matrix and conductive fillers (ATCBn,ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25

Ratio of PP matrix and additives	Trial1	Trial2	Trial3	Trial4	Trial5	Average	S.D.
	(S/cm)	(S/cm)	(S/cm)	(S/cm)	(S/cm)	(S/cm)	
PP:ATAB(50:50)	32.65	32.67	31.99	31.67	35.12	32.82	1.354
PP:ATCBn(50:50)	31.10	31.68	30.07	30.65	31.35	30.97	0.625
PP:ATGT(50:50)	26.86	26.33	26.35	27.34	25.12	26.40	0.827
PP:ATCbe(50:50)	36.49	36.08	35.23	35.11	39.55	36.49	1.802
PP:ATAB:ATCBn (50:25:25)	34.07	35.62	35.67	33.68	32.07	34.22	1.502
PP:ATAB:ATGT (50:25:25)	33.52	33.46	32.08	33.59	34.56	33.44	1.717
PP:ATAB:ATCbe (50:25:25)	39.32	39.65	38.24	38.24	40.15	39.12	0.855
PP:ATCBn:ATGT (50:25:25)	28.65	27.08	27.66	28.02	31.99	28.68	1.936
PP:ATCBn:ATCbe (50:25:25)	32.61	32.66	31.64	33.12	30.01	32.01	1.240
PP:ATGT:ATCbe (50:25:25)	30.22	30.65	31.24	30.31	28.68	30.22	0.950

Table C-3 Interfacial Contact Resistance (ICR by Through-plane technique of Polypropylene matrix and conductive fillers (ATCBn,ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25

Ratio of PP matrix and additives	ICR in different moment torque						
	60 lbf/ins	80 lbf/ins	100 lbf/ins	120 lbf/ins	140 lbf/ins	160 lbf/ins	180 lbf/ins
PP:ATAB(50:50)	12.90	11.91	11.01	10.17	9.64	9.24	8.85
PP:ATCBn(50:50)	14.09	13.02	12.03	11.11	10.54	10.10	9.68
PP:ATGT(50:50)	16.19	14.96	13.82	12.77	12.11	11.60	11.12
PP:ATCbe(50:50)	11.43	10.56	9.76	9.01	8.55	8.19	7.85
PP:ATAB:ATCBn (50:25:25)	12.17	11.25	10.39	9.60	9.11	8.72	8.36
PP:ATAB:ATGT (50:25:25)	13.16	12.15	11.23	10.37	9.84	9.42	9.03
PP:ATAB:ATCbe (50:25:25)	10.82	9.99	9.23	8.53	8.09	7.75	7.43
PP:ATCBn:ATGT (50:25:25)	14.95	13.81	12.76	11.79	11.18	10.71	10.26
PP:ATCBn:ATCbe (50:25:25)	13.29	12.23	11.30	10.44	9.91	9.49	9.09
PP:ATGT:ATCbe (50:25:25)	14.90	13.77	12.72	11.75	11.15	10.68	10.23

Table C-4 The density of Polypropylene and conductive fillers (ATCBn,ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25

Ratio of PP matrix and additives	Trial 1 Density (g/cm3)	Trial 2 Density (g/cm3)	Trial 3 Density (g/cm3)	Trial 4 Density (g/cm3)	Density average (g/cm3)	S.D
PP:ATAB(50:50)	1.18	1.23	1.22	1.17	1.20	0.029
PP:ATCBn(50:50)	1.42	1.38	1.39	1.45	1.41	0.032
PP:ATGT(50:50)	1.18	1.17	1.14	1.11	1.15	0.032
PP:ATCbe(50:50)	1.18	1.2	1.17	1.17	1.18	0.014
PP:ATAB:ATCBn(50:25:25)	1.46	1.48	1.44	1.50	1.47	0.026
PP:ATAB:ATGT(50:25:25)	1.21	1.24	1.25	1.22	1.23	0.018
PP:ATAB:ATCbe(50:25:25)	1.35	1.34	1.31	1.32	1.33	0.018
PP:ATCBn:ATGT(50:25:25)	1.37	1.34	1.35	1.30	1.34	0.029
PP:ATCBn:ATCbe(50:25:25)	1.15	1.13	1.1	1.14	1.13	0.022
PP:ATGT:ATCbe(50:25:25)	1.12	1.1	1.14	1.04	1.10	0.043

Table C- 5 Water absorption of Polypropylene and conductive fillers (ATCBn,ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25

Ratio of PP matrix and additives	Trial 1	Trial 2	Trial 3	Trial 4	Average Water absorption (%)	S.D
	Water absorption (%)	Water absorption (%)	Water absorption (%)	Water absorption (%)		
PP:ATAB(50:50)	0.081	0.084	0.085	0.082	0.083	0.002
PP:ATCBn(50:50)	0.074	0.077	0.079	0.074	0.076	0.002
PP:ATGT(50:50)	0.084	0.084	0.089	0.091	0.087	0.004
PP:ATCbe(50:50)	0.061	0.067	0.062	0.062	0.063	0.003
PP:ATAB:ATCBn (50:25:25)	0.095	0.099	0.093	0.101	0.097	0.004
PP:ATAB:ATGT (50:25:25)	0.101	0.107	0.103	0.109	0.105	0.004
PP:ATAB:ATCbe (50:25:25)	0.085	0.089	0.092	0.090	0.089	0.003
PP:ATCBn:ATGT (50:25:25)	0.077	0.081	0.079	0.075	0.078	0.003
PP:ATCBn:ATCbe (50:25:25)	0.065	0.069	0.064	0.070	0.067	0.003
PP:ATGT:ATCbe (50:25:25)	0.067	0.069	0.065	0.071	0.068	0.003

Table C-6 Hardness of Polypropylene and conductive fillers (ATCBn,ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25

Ratio of PP matrix and additives	Trial1	Trial2	Trial3	Trial4	Trial5	Average	S.D.
	HRD	HRD	HRD	HRD	HRD	HRD	
PP:ATAB(50:50)	75	75	74.5	74.5	73.5	74.5	0.612
PP:ATCBn(50:50)	72.5	73	72.5	72	70	72	1.173
PP:ATGT(50:50)	74.5	74.5	74	74	73	74	0.612
PP:ATCbe(50:50)	72	71.5	71	72	71	71.5	0.500
PP:ATAB:ATCBn (50:25:25)	75	75.5	75	76	76	75.5	0.500
PP:ATAB:ATGT (50:25:25)	77	77.5	77	77.5	76	77	0.612
PP:ATAB:ATCbe (50:25:25)	75	75.5	76	75	76	75.5	0.500
PP:ATCBn:ATGT (50:25:25)	74.5	74.5	75	75	73.5	74.5	0.612
PP:ATCBn:ATCbe (50:25:25)	73	73.5	73.5	73	72	73	0.612
PP:ATGT:ATCbe (50:25:25)	74.5	75	75	74.5	76	75	0.612

Table C-7 The flexural strength of the polypropylene with conductive fillers (ATCBn,ATAB,ATGT,ATCBe) in ratio of 50:50 and 50:25:25

Ratio of PP matrix and additives	Trial1	Trial2	Trial3	Trial4	Trial5	Average	S.D.
	Flexural Strength (Mpa)	Flexural Strength (Mpa)	Flexural Strength (Mpa)	Flexural Strength (Mpa)	Flexural Strength (Mpa)	Flexural Strength (Mpa)	
PP:ATAB(50:50)	21.22	21.35	22.35	20.58	20.61	21.22	0.718
PP:ATCBn(50:50)	19.85	20.41	19.64	20.35	20.19	20.09	0.332
PP:ATGT(50:50)	24.73	24.26	23.66	24.22	26.74	24.72	1.190
PP:ATCbe(50:50)	18.01	18.77	19.66	19.22	19.60	19.05	0.686
PP:ATAB:ATCBn (50:25:25)	25.88	24.35	23.52	24.66	25.90	24.86	1.028
PP:ATAB:ATGT (50:25:25)	25.12	26.85	27.00	26.55	26.66	26.44	0.754
PP:ATAB:ATCbe (50:25:25)	22.35	22.15	23.47	22.11	21.99	22.41	0.603
PP:ATCBn:ATGT (50:25:25)	25.85	25.31	25.97	24.38	26.61	25.62	0.837
PP:ATCBn:ATCbe (50:25:25)	21.37	21.05	21.32	21.35	20.19	21.06	0.500
PP:ATGT:ATCbe (50:25:25)	25.27	25.31	25.15	24.66	26.51	25.38	0.684

Appendix D

Study of the effect of mixture of carbon nanotubes

Table D-1 Electrical conductivity by in-plane technique of Polypropylene matrix and conductive fillers (ATAB:ATCBe) with CNTs (Timesnano) and CNT (N7000)

Ratio of PP matrix and additives	Trial1	Trial2	Trial3	Trial4	Trial5	Average	S.D.
	(S/cm)	(S/cm)	(S/cm)	(S/cm)	(S/cm)	(S/cm)	
PP:ATAB:ATCBe (50:25:25)	42.50	42.10	42.51	42.46	40.79	42.07	0.74
PP:ATAB:ATCBe:CNTs (N7000) (50:24:24:2)	46.95	47.28	47.00	46.89	47.37	47.10	0.21
PP:ATAB:ATCBe:CNTs (N7000) (50:23.5:23.5:3)	48.78	49.32	48.72	49.32	49.31	49.09	0.31
PP:ATAB:ATCBe:CNTs (N7000) (50:23:23:4)	50.76	50.31	50.44	50.57	50.46	50.51	0.17
PP:ATAB:ATCBe:CNTs (Timesnano) (50:24:24:2)	46.40	46.30	46.08	46.03	46.34	46.23	0.16
PP:ATAB:ATCBe:CNTs (Timesnano) (50:23.5:23.5:3)	46.35	46.73	46.84	46.51	46.47	46.58	0.20
PP:ATAB:ATCBe:CNTs (Timesnano) (50:23:23:4)	49.88	49.32	49.44	49.38	49.53	49.51	0.22

Table D-2 Electrical conductivity by Through-plane technique of Polypropylene matrix and conductive fillers (ATAB:ATCBe) with CNTs (Timesnano) and CNT (N7000)

Ratio of PP matrix and additives	Trial1	Trial2	Trial3	Trial4	Trial5	Average	S.D.
	(S/cm)	(S/cm)	(S/cm)	(S/cm)	(S/cm)	(S/cm)	
PP:ATAB:ATCBe (50:25:25)	39.32	39.65	38.24	38.24	40.15	39.12	0.855
PP:ATAB:ATCBe:CNTs (N7000) (50:24:24:2)	43.51	42.96	43.65	44.25	43.68	43.61	0.461
PP:ATAB:ATCBe:CNTs (N7000) (50:23.5:23.5:3)	45.98	45.63	44.39	46.31	47.15	45.89	1.011
PP:ATAB:ATCBe:CNTs (N7000) (50:23:23:4)	47.25	47.63	47.06	48.05	45.62	47.12	0.919
PP:ATAB:ATCBe:CNTs (Timesnano) (50:24:24:2)	41.06	42.86	43.27	41.10	43.62	42.38	1.218
PP:ATAB:ATCBe:CNTs (Timesnano) (50:23.5:23.5:3)	44.09	44.32	43.69	43.26	45.25	44.12	0.747
PP:ATAB:ATCBe:CNTs (Timesnano) (50:23:23:4)	45.49	45.07	45.61	46.36	46.17	45.74	0.525

Table D-3 Interfacial Contact Resistance (ICR by Through-plane technique of Polypropylene matrix and conductive fillers (ATAB:ATCBe) with CNTs (Timesnano) and CNT (N7000)

Ratio of PP with additives	ICR in different moment torque						
	60	80	100	120	140	160	180
PP:ATAB:ATCBe:CNTs(N7000) (50:24:24:2)	8.98	8.30	7.67	7.08	6.72	6.43	6.17
PP:ATAB:ATCBe:CNTs(N7000) (50:23.5:23.5:3)	8.35	7.71	7.13	6.58	6.24	5.98	5.73
PP:ATAB:ATCBe:CNTs(N7000) (50:23:23:4)	7.47	6.90	6.38	5.89	5.59	5.35	5.13
PP:ATAB:ATCBe:CNTs(Timesnano) (50:24:24:2)	9.27	8.56	7.91	7.31	6.93	6.64	6.36
PP:ATAB:ATCBe:CNTs(Timesnano) (50:23.5:23.5:3)	9.16	8.46	7.81	7.22	6.85	6.56	6.29
PP:ATAB:ATCBe:CNTs(Timesnano) (50:23:23:4)	8.22	7.59	7.01	6.48	6.15	5.89	5.64

Table D-4 Density of Polypropylene matrix and conductive fillers (ATAB:ATCBe) with CNTs (Timesnano) and CNT (N7000)

Ratio of PP matrix and additives	Trial 1	Trial 2	Trial 3	Trial 4	Density average (g/cm3)	S.D
	Density (g/cm3)	Density (g/cm3)	Density (g/cm3)	Density (g/cm3)		
PP:ATAB:ATCbe (50:25:25)	1.35	1.34	1.31	1.32	1.33	0.018
PP:ATAB:ATCBe:CNTs (N7000) (50:24:24:2)	1.32	1.35	1.31	1.34	1.33	0.018
PP:ATAB:ATCBe:CNTs (N7000) (50:23.5:23.5:3)	1.36	1.32	1.3	1.42	1.35	0.053
PP:ATAB:ATCBe:CNTs (N7000) (50:23:23:4)	1.37	1.38	1.35	1.42	1.38	0.029
PP:ATAB:ATCBe:CNTs (Timesnano) (50:24:24:2)	1.31	1.26	1.28	1.31	1.29	0.024
PP:ATAB:ATCBe:CNTs (Timesnano) (50:23.5:23.5:3)	1.32	1.35	1.37	1.32	1.34	0.024
PP:ATAB:ATCBe:CNTs (Timesnano) (50:23:23:4)	1.36	1.39	1.35	1.34	1.36	0.022

Table D-5 Water absorbtion of Polypropylene matrix and conductive fillers (ATAB:ATCBe) with CNTs (Timesnano) and CNT (N7000)

Ratio of PP matrix and additives	Trial 1	Trial 2	Trial 3	Trial 4	Average	S.D
	Water absorption (%)	Water absorption (%)	Water absorption (%)	Water absorption (%)	Water absorption (%)	
PP:ATAB:ATCbe (50:25:25)	0.085	0.089	0.092	0.090	0.089	0.003
PP:ATAB:ATCBe:CNTs (N7000) (50:24:24:2)	0.106	0.109	0.107	0.110	0.108	0.002
PP:ATAB:ATCBe:CNTs (N7000) (50:23.5:23.5:3)	0.117	0.118	0.116	0.121	0.118	0.002
PP:ATAB:ATCBe:CNTs (N7000) (50:23:23:4)	0.12	0.123	0.122	0.119	0.121	0.002
PP:ATAB:ATCBe:CNTs (Timesnano) (50:24:24:2)	0.087	0.088	0.09	0.091	0.089	0.002
PP:ATAB:ATCBe:CNTs (Timesnano) (50:23.5:23.5:3)	0.091	0.094	0.09	0.093	0.092	0.002
PP:ATAB:ATCBe:CNTs (Timesnano) (50:23:23:4)	0.099	0.1	0.103	0.102	0.101	0.002

Table D-6 the hardness of PP:ATAB:ATCBe with 2-4 % of fillers CNTs (Timesnano) and CNT (N7000)

Ratio of PP matrix and additives	Trial1	Trial2	Trial3	Trial4	Trial5	Average	S.D.
	HRD	HRD	HRD	HRD	HRD	HRD	
PP:ATAB:ATCbe (50:25:25)	75	75.5	76	75	76	75.5	0.500
PP:ATAB:ATCBe:CNTs (N7000) (50:24:24:2)	77	77.5	78	77.5	80	78	1.173
PP:ATAB:ATCBe:CNTs (N7000) (50:23.5:23.5:3)	78.5	79	78	79	80.5	79	0.935
PP:ATAB:ATCBe:CNTs (N7000) (50:23:23:4)	79	78.5	79	78	83	79.5	2.000
PP:ATAB:ATCBe:CNTs (Timesnano) (50:24:24:2)	77	77.5	77.5	77	78.5	77.5	0.612
PP:ATAB:ATCBe:CNTs (Timesnano) (50:23.5:23.5:3)	78.5	78	78	79	76.5	78	0.935
PP:ATAB:ATCBe:CNTs (Timesnano) (50:23:23:4)	79	78.5	78.5	79	80	79	0.612

Table D-7 The flexural strength of the Polypropylene matrix and conductive fillers (ATAB:ATCBe) in ratio of 50:25:25 with CNTs (Timesnano) and CNT (N7000)

Ratio of PP matrix and additives	Trial1	Trial2	Trial3	Trial4	Trial5	Average	S.D.
	Flexural Strength (Mpa)	Flexural Strength (Mpa)	Flexural Strength (Mpa)	Flexural Strength (Mpa)	Flexural Strength (Mpa)	Flexural Strength (Mpa)	
PP:ATAB:ATCbe (50:25:25)	22.35	22.15	23.47	22.11	21.99	22.41	0.603
PP:ATAB:ATCBe:CNTs (N7000) (50:24:24:2)	24.02	24.24	23.22	23.45	25.79	24.15	1.010
PP:ATAB:ATCBe:CNTs (N7000) (50:23.5:23.5:3)	23.52	23.84	24.06	24.08	27.15	24.53	1.483
PP:ATAB:ATCBe:CNTs (N7000) (50:23:23:4)	25.36	25.66	25.18	24.33	26.21	25.35	0.688
PP:ATAB:ATCBe:CNTs (Timesnano) (50:24:24:2)	24.35	24.10	23.14	23.12	25.94	24.13	1.153
PP:ATAB:ATCBe:CNTs (Timesnano) (50:23.5:23.5:3)	24.46	24.65	25.31	23.35	26.60	24.88	1.197
PP:ATAB:ATCBe:CNTs (Timesnano) (50:23:23:4)	24.62	25.24	25.65	25.31	24.80	25.12	0.413

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