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Properties of Epoxy/Carbon Black/Graphite Composites for Bipolar Plate in Polymer Electrolyte Membrane Fuel Cell

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Abstract. Epoxy resin (EP) composites including carbon black (CB) and graphite (G) were produced and investigate. The in-plane electrical conductivity of EP/CB/G composites was measured. Various weight percentages (wt.%) of CB as a secondary filler and G as a primary filler were added into the EP as a matrix. Dispersion of CB and G within matrix were conducted by an internal mixer (Haake Reomix). Mixture of EP/CB/G was poured into the steel mold, and produced through hot press machine. In-plane electrical conductivity of EP/CB/G composites in terms of variation of CB and G concentration were measured by the Jandel multi high four point probe. The highest in-plane electrical conductivity of the EP/CB/G composites obtained was 120 S/cm. This value exceeds the requirement of U.S Department of Energy (DOE) target for bipolar plate application (> 100 S/cm).

Introduction

Polymer electrolyte membrane (PEM) fuel cell is an interesting subject for researches from alternative energy and environmental considerations due to their attractive features in high power density and low operating temperature [1, 2]. However, on the fundamental of commercial manufacture, it is related with some major problems such as high fabrication cost and not enough reliability. The bipolar plate is an important component in PEM fuel cell, which can contribute around 80 % of total weight and 45 % of stack cost [3]. Hence, the investigation of bipolar plates for cost and performance materials has become a significant research issues. The conventional materials for fabricating bipolar plate are based on graphite materials or metals. The most commonly use bipolar plate material is graphite plates for some advantages of high electrical conductivity, lower density than those of metals and excellent corrosion resistance. However, some disadvantages of graphite plates including the high cost resulted from machining channels into the surface and their brittleness would cause the fuel cell stack to be heavy and huge [4, 5]. Therefore, graphite polymer composite bipolar plates has been considered as the alternate for the conventional graphite plates, and then indicated as lower cost, higher flexibility, lighter weight and easier developed. The crucial problem of the polymer composite bipolar plate is the low electrical conductivity. Graphite based polymer composite bipolar plates has been made-up from the incorporation of high loading concentration of graphite and additional conductive fillers to improve the electrical conductivity [4-6]. Usually conductive fillers used consist of carbon nanotube (CNTs), carbon fiber (CF), and carbon black (CB) which have been incorporated into the composites to increase the performance of composite bipolar plate through conventional polymer processing [6, 7]. Although the significant improvement in the electrical conductivity was reached, a very high conductive fillers is still required to meet the electrical conductivity of bipolar plate by the U.S Department of Energy (DOE) targets (>100 S/cm) [8]. Wolf et al has developed a liquid crystal polymer composite bipolar plate using compression molding method [9]. The conductive fillers

were a combination of CB and CF, and loading concentration level in the composite was below 40 vol. %. However, electrical conductivity obtained was less than 6 S/cm although the bipolar plate had an excellent gas barrier and mechanical properties. Dweiri and Sahari studied CB/G/PP composite bipolar plates by compression molding process [10]. The electrical conductivity obtained was only around 35 S/cm although graphite was added as high as 80 wt. %. Similar research had also done by Yin et al. very high conductive filler of 85 wt. % graphite powder was required to reach U.S DOE targets of 100 S/cm [11]. In this study, epoxy based composites contained conductive fillers of CB as secondary filler and graphite as primary filler were developed to enhance the electrical conductivity of EP/CB/G composites in order to achieve the requirement of the U.S DOE targets for bipolar plate application. The in-plane electrical conductivity and flexural strength of EP/CB/G composites were investigated.

Experimental

Materials. Two conductive fillers with different sizes and shapes were used in this study, Graphite (G) type 4012 has surface area of 1.5 m^2/g, average particle size of 74 μm and purity is ≥ 99 % as reported by manufacturer. The G was purchased from Insutex Industries Sdn. Bhd., Malaysia as the local agent for Asbury Carbons Inc., U.S. The carbon black (CB) type 5303 were also purchased from Insutex Industries Sdn. Bhd., Malaysia, with a surface area of 254 m^2/g, average particle size 30 nm, and a purity is ≥ 99% as reported by manufacturer. The epoxy resin was a bisphenol-A based epoxy resin, with a viscosity 6 Poise. The curing agent was a diamine type (tetra functional) to facilitate rapid and cross-linking in the epoxy resin. The epoxy resin and curing agent were purchased from Mid Western Lab Suppliers., Malaysia as the local agent for U.S Composites.

Fabrication of EP/CB/G composites. The liquid epoxy resin and curing agent were mixed with weight ratio 3:1 as recommended by manufacturer. The mixing of CB/G in epoxy was produced in some stages. First, CB and G were mixed with different wt. % using ball milling to get the homogenous mixture. The weight ratio of ball and powder were 4:1 with stainless steel balls (10 mm in diameter) at rotating speed of 200 rpm for one hour. EP and curing agent then were mixed using the high speed mechanical mixer (RW 20-KIKA-WERK) at 1200 rpm for 40 seconds as the second stage. Lastly, the mixture of EP/CB/G further mixed using the internal mixer (Haake Reomix) at temperature 30°C. The rotational speed and the mixing time were set at 25 rpm and 10 min, respectively [12]. The various composition of G, CB and EP were 50, 55, 60 wt. %, 20, 25, 30 wt. % and 20 wt. %, respectively. The composite mixtures were then poured into a steel mould at molding temperature 150 °C and molding pressure 30 MPa were applied for 1.5 hours [13].

In-plane electrical conductivity of EP/CB/G composites. The in-plane electrical conductivity of the EP/CB/G composites was measured using a Jandel Multi Height Four Point Probe combined with RM3 Test Units which has constant current source and digital voltmeter designed especially for the four point probe measurement. The system accuracy was within 0.3 % [10].

Flexural strength EP/CB/G composites. The flexural strength of the EP/CB/G composites were measured by three point bending according to ASTM D790-03 at room temperatur using Universal Testing Machine Model Instron 5567 at cross head speed of 1 mm/min. The dimensions of the specimens were 100×12.7×2.5 mm, and support span length of each specimen was fixed at 50.8 mm.

Morphology. Surface morphology of the all specimens were observed using Olympus microscope.

Results and Discussion

In-plane electrical conductivity of EP/CB/G composites. The effect of addition of carbon black (CB) as the second fillers in the G/epoxy composites shown in Figure1. The in-plane electrical conductivity of the EP/CB/G composites increase rapidly (50%) with the increasing of CB content from 20 wt. % to 25 wt. %. Addition 25 wt. % of CB shows the highest in-plane electrical conductivity of EP/CB/G composites as 120 S/cm. High surface area (254 m^2/g) and a very small diameter (30 nm) of CB particles within epoxy resin act as a bridge between flake-like graphite and

build more conductive pathway network throughout the EP/CB/G composites. It is enhancing the in-plane electrical conductivity of the EP/CB/G composites up to 25 wt. % CB addition. Nevertheless, more addition of CB as a secondary filler decreased the in-plane electrical conductivity because the epoxy resin as a matrix is not sufficient enough to bind the filler more than 25 wt. %. Mighri et al. explained the same fenomenon as a wetting limitation that decreased the electrical conductivity of the EP/CB/G composites [14]. Dweiri and Sahari reported that the electrical conductivity of PP/CB/G was only around 35 S/cm on composition 20/25/55 [10]. Compared to this study, on the same weight percentage of CB (25 wt. %) as a secondary filler, shows higher in-plane electrical conductivity values than Dweiri and Sahari [10]. The different result of the in-plane electrical conductivity was because of the absent from any polar group in polypropylene (PP) back bone that makes the dispersion of CB as a secondary filler within the polymer matrix more difficult [15].

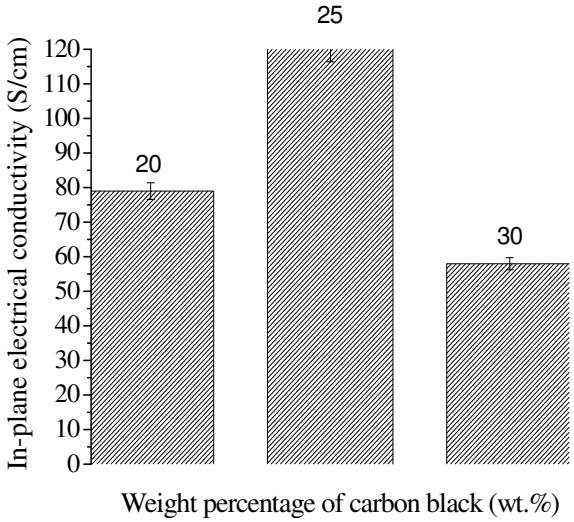

Fig.1. In-plane electrical conductivity of EP/CB/G composites of (20/20/60), (20/25/55) and (20/30/50)

Flexural strength of EP/CB/G/ composites. Effect of addition CB as secondary filler on the flexural strength of the EP/CB/G composites shows in Figure 2.

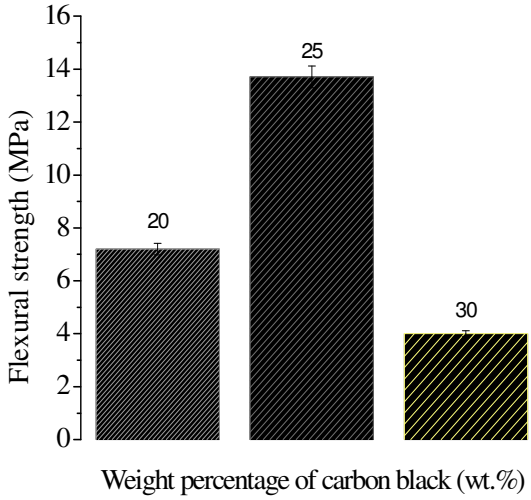

Fig.2. Flexural strength of EP/CB/G composites of (20/20/60), (20/25/55) and (20/30/50)

Figure 2. Shows that the addition of CB 25 wt.% increase the flexural strength sharply from 7 MPa to 14 MPa, however the flexural strength decrease with addition of CB on 30 wt.%. The value of the flexural strength of EP/CB/G composites shows a similar trend with in-plane electrical conductivity behavior. High aspect ratio of CB as the secondary filler and good interfacial adhesion between CB and matrix are also contributed to enhance flexural strength of EP/CB/G composite. Lack of binding to the conductive fillers due to fabrication process produced defect of the composite structure and decrease the flexural strength in high CB content (above 25 wt.%) [16, 17].

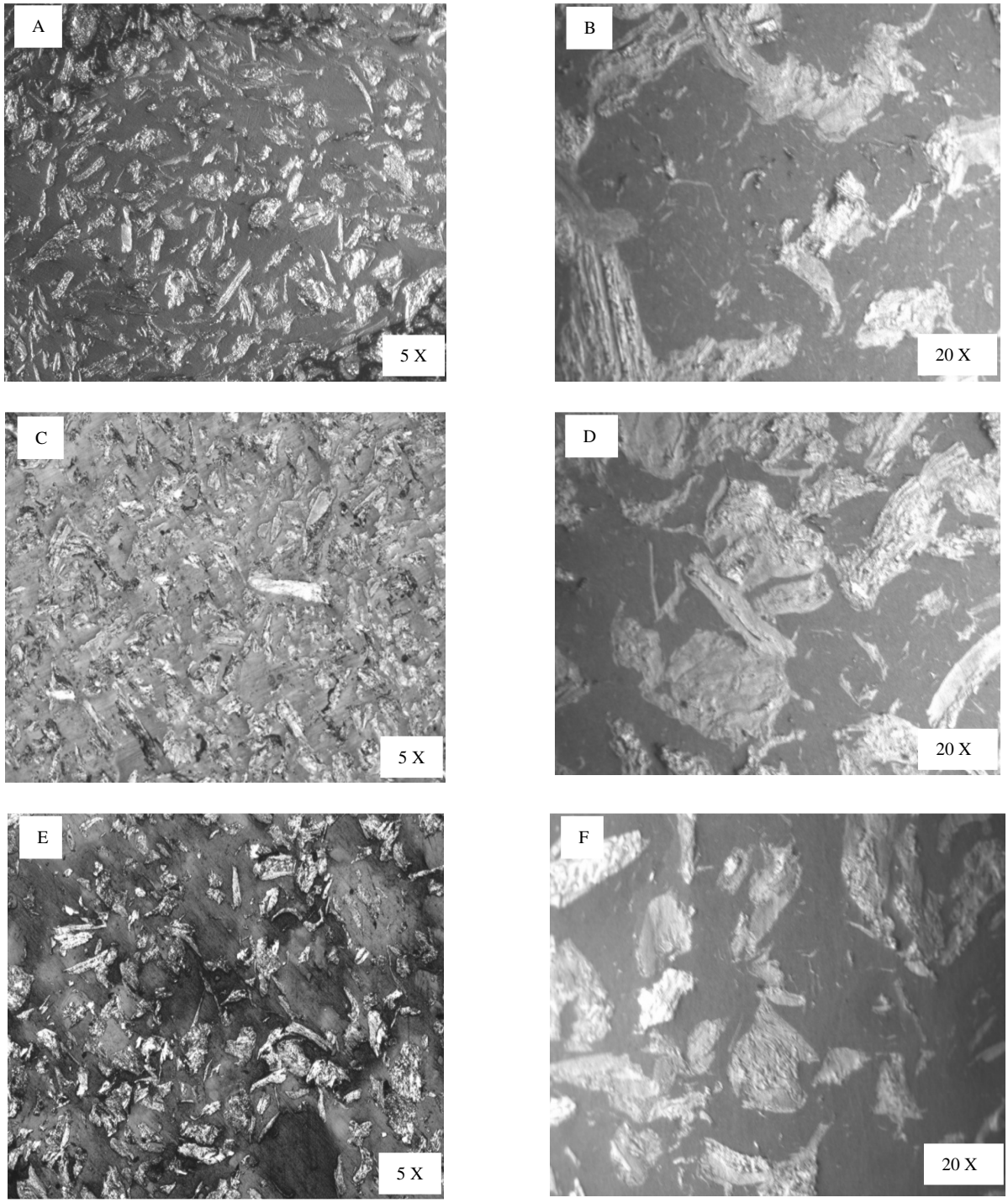

Fig.3.Optical microscope surface of EP/CB/G composites of (a,b) 20/20/60, (c,d) 20/25/55, (e,f) 20/30/50 (in wt. %) with different magnification of a,c,e) 5X and, b,d,f) 20X

The optical microscope surfaces of EP/CB/G composites are shown in Figure 3. The bright images indicate as G and the dark as CB. G and CB are reasonably well dispersed and distributed over the whole area and even between the graphite particles at 20 and 25 wt. % of CB. However, G agglomeration can be observed with the increasing of CB content (30 wt. %).

Summary

The EP/CB/G composites with various weight percentages (wt.%) of conductive fillers have been fabricated using hot press machine. Some findings are:

1. The highest in-plane electrical conductivity of EP/CB/G composites obtained was 120 S/cm, this value exceeds the requirement of bipolar plate by U.S Department of Energy (DOE).
2. The decreasing of the in-plane electrical conductivity and flexural strength of EP/CB/G composites at 25 wt.% of CB can be explained by the formation of graphite agglomeration and large distance conductive fillers (G and CB) particles, which degrades the in-plane electrical conductivity and flexural strength of EP/CB/G composites.

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