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Effect of Formation Temperature on Properties of Graphite/Stannum Composite for Bipolar Plate

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Abstract. Bipolar plates are key components in Proton Exchange Membrane (PEM) fuel cells. They carry current away from the cell and withstand the clamping force of the stack assembly. Therefore, PEM fuel cell bipolar plates must have high electrical conductivity and adequate mechanical strength, in addition to being light weight and low cost in terms of both applicable materials and production methods. In this research, the raw materials used to fabricate the high performance bipolar plate are Graphite (Gr), Stannum (Sn) and Polypropylene (PP). All materials used was in powder form and Gr and Sn act as fillers and the PP acts as binder. The ratio of fillers (Gr/Sn) and binder (PP) was fixed at 80:20. For the multi-conductive filler, small amount of Sn, which is 10 up to 20wt% (from the total weight of fillers 80%) have been added into Gr/Sn/PP composite. The fillers were mixed by using the ball mill machine. The second stage of mixing process between the mixer of fillers and binder is also carried out by using ball mill machine before the compaction process by the hot press machine. The effect of formation temperatures (160°C-170°C) on the properties of Gr/Sn/PP composite had been studied in detail, especially the electrical conductivity, bulk density, hardness and microstructure analysis of Gr/Sn/PP composite. The result shows that there are significant improvement in the electrical conductivity and bulk density, which are exceeding the US-DoE target with the maximum value of 265.35 S/cm and 1.682g/cm^3 respectively.

INTRODUCTION

Fuel cell is an electrochemical energy conversion device which can convert the chemical energy from hydrogen and oxygen into water, heat and in the process it will produce electricity. Hydrogen is commonly acts as fuel and sometimes hydrocarbons such as natural gas and alcohols like methanol are used. Fuel Cells generate electricity through an electrochemical process in which the energy stored in a fuel is converted directly into electricity. Fuel cells are extremely attractive from an environmental standpoint because electrical energy is generated without combusting fuel. Fuel cells are known as great potential to be low emission power generation sources in the future due to its attractive characteristics such as high energy conversion efficiency [1], very low chemical and acoustical pollution [2]. Unlike power sources that use fossil fuels, the by-products from an operating fuel cell are heat and water. In the future, fuel cells are known as the most potential power sources for residential, mobile and automotive applications. Fuel cells have the potential to replace the internal combustion engine in vehicles and provide power in stationary and portable power applications because they are energy efficient, clean, and fuel flexible [3-5].

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According to Ahmad, et al. (2013), the performance of Polymer Exchange Membrane Fuel Cell (PEMFC) in producing energy is depending on the bipolar plate materials used. The problem that faced by researchers is the properties of carbon-based materials which have poor electrical and thermal conductivity, fragile structure and low mechanical strength [6]. However, other materials such as metal has high tendency to corrosion, costly and time consuming for machining [7-8]. In order to tackle the weaknesses of pure Gr as bipolar plate, the hybrid conductive composite (HCC) material was chosen as bipolar plate becomes preferable [6]. The Gr/Sn/PP composite bipolar plate can be easily manufactured by the hot compression molding method and based on previous research that can be applied as bipolar plate in PEMFC [9]. In this research the ratio of fillers (Gr/Sn) and binder (PP) when it is 80:20 will give the best result in the test such as flexure test, density test, hardness and microstructure analysis, but the problem faced by the researchers is the electrical conductivity of the bipolar plate is too low and can't achieve the US-DoE target as a PEMFC bipolar plate. The aim of this research is to study the effect of formation temperatures on the properties of Gr/Sn/PP composite.

EXPERIMENTAL PROCEDURE

Materials

The materials used in this research are Gr and Sn as conductive fillers and PP as a binder. The property requirements shown in Table 1 should be satisfied for the fabrication of a bipolar plate. Meanwhile Table 1 shown the comparison of properties between Gr, Sn and PP.

TABLE 1. The Characteristics of the Materials

Materials	**Gr**	**Sn**	**PP**
Grade	3243	Aldrich	Titan(600)
Density g/cm^3	1.74	7.31	0.91-0.92
Thermal Stability °C	3500-4000	210-240	175-220
Size µm	65.901	33.822	250
Shape	Flake	Particulate	Particulate
Resistivity Ωcm	1295(10^{-8})	115(10^{-9})	10^{14}

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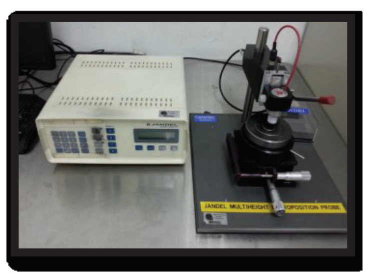
(a)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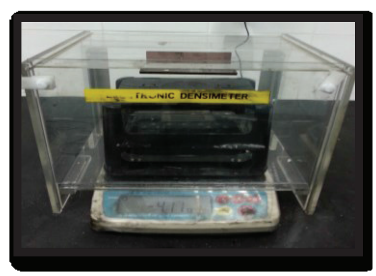
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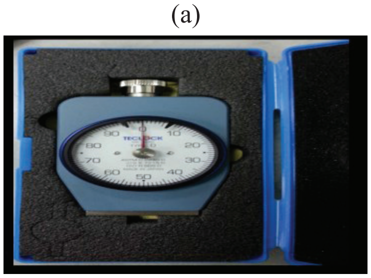
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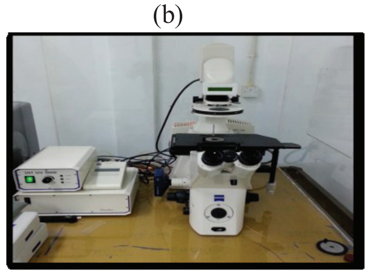
(e)

FIGURE 1. (a) Hot Press Machine, (b) Jandel Multi Height Four Points Probes, (c) Electronic Densimeter, (d) Type-D Analog Shore Hardness Tester and (e) Confocal Inverted Microscope.

Preparation of Composites

The ratio of fillers and matrix has been fixed 80:20. For initial study from 80% fillers, the Sn loading has been varied from 5 wt% up to 30 wt%. But for this sample, PP has been used in granular form. Before the fabrication process, all filler was mixed used ball mill machine for a half hours. After that the mixed fillers has been mixed with PP by using internal mixer machine at temperature of 200 ^{0}C, torque 50 Nm and duration around 17 minutes. The pulverizing process was done to produce the fine particles of compound G/Sn/PP before the fabrication process of specimens was produced by using hot press machine as shown in Fig. 1 (a) and mold with the size of 50 mm x 50 mm at temperature of 175 ^{0}C for 10 min with pressure of 15 tons.

Formation Temperatures

Three loading ratios of Gr/Sn composite were prepared by 70/10, 65/15 and 60/20 respectively. These compositions were mixed by using ball mill machine for one and a half hours. After mixing process, the fabrication process of specimens was produced by using hot press machine and mold with the size of 50 mm x 50 mm at temperatures of 160, 165 and 170 0C for 10 min with pressure of 15 tons.

Test Method

The effect of Sn on the properties of Gr/Sn composites such as electrical conductivity, hardness, bulk density and microstructure was investigated. For electrical conductivity test, Jandel Multi Height Four Points Probes were used, hardness test was performed by using Digital Shore Scale D type Durometer according to ASTM D2240 and densimeter was used to investigate bulk density. Finally, microstructure test was carried out using Confocal Microscope and Fig. 2 (b) up to (e) shown the equipment's has been used for the tests.

RESULTS AND DISCUSSION

Effect of Sn loading on electrical conductivity

The electrical conductivity of Gr/Sn/PP composites with Sn loading of 5% up to 30 wt% are shown in Fig.2. According to the graph, the electrical conductivity has increased with the increment of Sn loading until Sn loading 20%, and after that the electrical conductivity has been decreased.The highest electrical conductivity shows by Sn loading of 20 wt% (15.05 S/cm) and the lowest shows by Sn loading 0 wt% (8.78 S/cm) respectively. All samples have been pressed at the temperature of 175 ^{0}C and mixed used internal mixer at the temperature of 200 ^{0}C. The value of electrical conductivity, low are due to over melted of PP as binder and at this stage PP is only become binder between filler particle but covered all particles. This will effect the conducting tunnel among the Gr and Sn and gives more barrier to conducting the network.

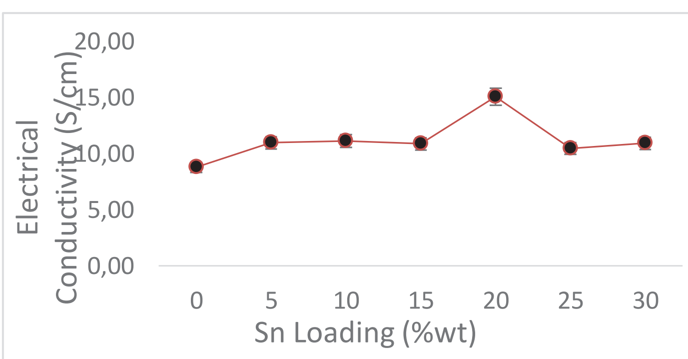

FIGURE 2. Electrical Conductivity (S/cm) versus Sn Content (wt%)

Effect of formation temperatures on electrical properties

Based on Sn loading and Fig. 2, the highest electrical conductivity is shown at 20 wt%. So that this composition (60/20/20) was used to determine the effect of formation temperatures. But all materials used was in powder form and the mixing process was used ball mill machine. During the fabrication process, the specimens were produced by using hot press machine and mold with the size of 50 mm x 50 mm. The temperatures were set at 160, 165 and 170 ^{0}C for 10 min with pressure of 15 tons. Figure 3 is shown the result of electrical conductivity at various temperatures. Based on this result, as temperature has been increased, the electrical conductivity also increased. The temperature of 170^{o}C was shown the highest value of electrical conductivity which is 50 S/cm^2. So that the temperature 170 ^{o}C was the most suitable temperature for fabrication specimens for future investigation of effect Sn loading.

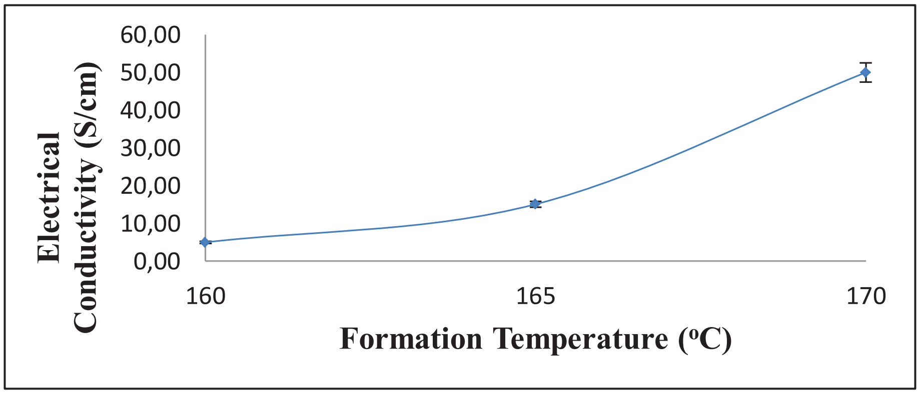

FIGURE 3. Graph of Electrical Conductivity (S/cm) against Formation Temperature (^{o}C)

Formation Temperature and Various Sn Loading

Several Sn loading has been selected which is 10, 15 and 20%wt and the temperature of 170 ^{0}C was used during fabrication process to produce the specimens. Fig.4 shows the electrical conductivity for each specimen at the formation temperature of 170^{o}C. Based on the result it is shown the drastic increment as compared with the value in Fig. 3. The electrical conductivity value of specimens has been prepared with used all materials in powder form and formation temperature of 170 ^{0}C had reached the requirement of the US DoE, where the electrical conductivity should be more than 100 S/cm. All specimens have shown the electrical conductivity is nearly or above 100 S/cm and the specimen with Sn 20wt% has shown the highest value of 265 S/cm. The result also shows that not only the Sn loading, but also the formation temperature affected the result of electrical conductivity.

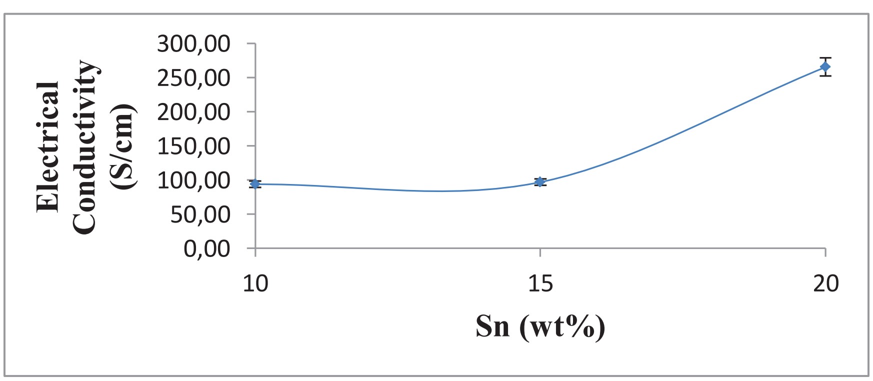

FIGURE 4. Graph of Electrical Conductivity (S/cm) against Weight Percentage of Sn (wt%)

Bulk Density

According to the Fig. 5, the graph of the mean value of bulk density (g/cm^3) against the weight percentage of Sn (wt%) was plotted. It shows that the bulk density of the Gr/Sn/PP composite has increased proportionally with the increment of weight percentage of Sn (wt%). The highest value of bulk density of the Gr/Sn/PP composite is 1.682 g/cm^3at 20 wt% of Sn, this is due to the higher composition and density of Sn.

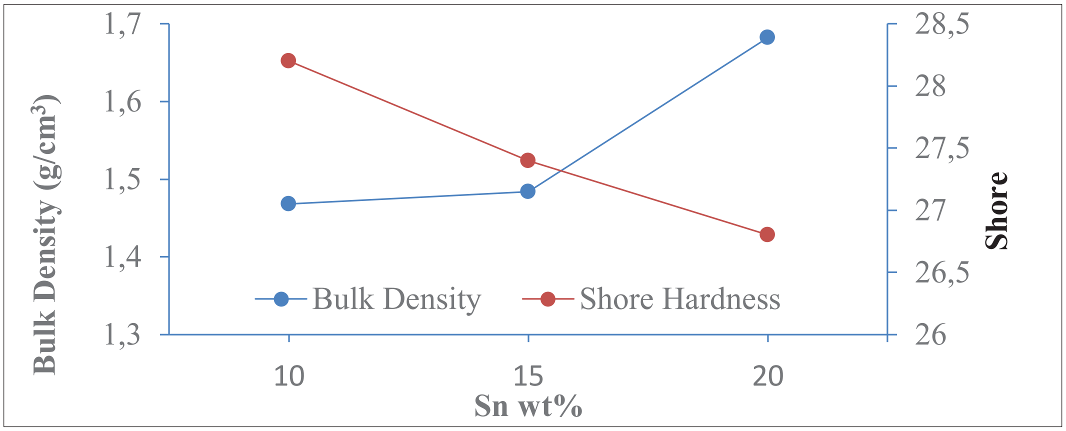

FIGURE 5. Graph of Bulk Density (g/cm^3) and Shore Hardness against Weight Percentage of Sn(wt%)

The bulk density is an important property for the Gr/Sn/PP composite for bipolar plate in PEMFC. Bulk density of specimens normally affected by the weight and size of particles. The lesser the weight of the filler materials, the lower the bulk density [4]. Figure 5 shows that the trend of bulk density with different composition of Sn. The bulk density increased with the increments composition of Sn.

Shore Hardness

According to the graph of the average value of shore hardness against the weight percentage of Sn (wt%) was plotted as shown in Fig. 5. The shore hardness of the Gr/Sn/PP bipolar plate does not show a constant trend with the increment of weight percentage of Sn (wt%) as shown in Fig. 5. The highest average value of shore hardness of the Gr/Sn/PP composite is 28.2 at 10 wt% of Sn. Shore hardness is the resistance of the materials towards indentation. In this research, all the specimens do not achieve the basic requirement of DoE which is 50. Figure 5 shows that the result of the shore hardness testing and the highest value for the shore hardness is 28.2 at the composition. The observation shows that the shore hardness was affected by the compaction of the materials and the voids that formed during fabrication process. The duration of cooling process of the samples will also give the significant effect on the shore hardness. The composite will behave brittle due to rapid cooling process.

Microstructure of Gr/Sn composites

Figure 6 showed the microstructure of Gr/Sn composites with various loadings of Sn. The microstructure shows that the dark area is the area rich with Gr and the bright area is the area rich with Sn [10]. As the Sn loading increased, the bright area has increased as compared to lower loading of Sn. The microstructure also shows the distribution of Sn particles has been distributed evenly. The distribution of Sn particles in each composite has significant effects on the properties of Gr/Sn composite. The electrical conductivity was increased that is due to conducting tunnel among the Gr and Sn and gives more conducting the network.

Gr/Sn/PP	70/10/20	65/15/20	60/20/20

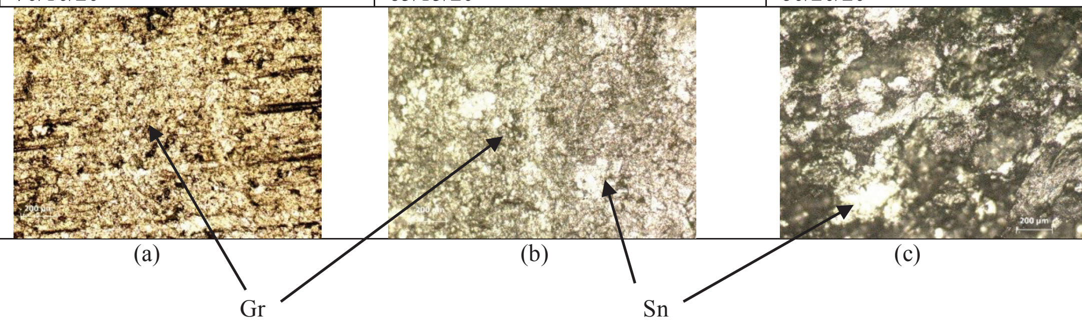

FIGURE 6. Microstructure of Gr/Sn/PP Composites (a) 10 wt% of Sn (b) 15% wt% of Sn and (c) 20wt% of Sn

CONCLUSION

The addition of Sn obviously has a significant effect on electrical conductivity and bulk density of Gr/Sn/PP composites. The addition of Sn in Gr composite offers a high opportunity in production of high conductive composites especially for PEMFC bipolar plates. Seem that the formation temperatures also have given significant effect to electrical conductivity and mechanical properties Gr/Sn/PP composite. The formation temperature of 170 ^{0}C with 20 wt% Sn was shown the highest electrical conductivity which is 265 S/cm. The bulk density and hardness are also decreased by addition of Sn loading. Meanwhile, further study on mechanical properties such as flexural strength and hardness should be pursued and make sure the value of this property meet the DoE target.

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