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Effect of the addition of milled carbon fiber as a secondary filler on the electrical conductivity of graphite/epoxy composites for electrical conductive material

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ABSTRACT

In this work, carbon composite bipolar plates consisting of synthetic graphite and milled carbon fibers as a conductive filler and epoxy as a polymer matrix developed using compression molding is described. The highest electrical conductivity obtained from the described material is 69.8 S/cm for the in-plane conductivity and 50.34 S/cm for the through-plane conductivity for the composite containing 2 wt.% carbon fiber (CF) with 80 wt.% filler loading. This value is 30% greater than the electrical conductivity of a typical graphite/epoxy composite with 80 wt.% filler loading, which is 53 S/cm for the in-plane conductivity and 40 S/cm for the through-plane conductivity. The flexural strength is increased to 36.28 MPa compared to a single filler system, which is approximately 25.22 MPa. This study also found that the General Effective Media (GEM) model was able to predict the in-plane and through-plane electrical conductivities for single filler and multiple filler composites.

1. Introduction

Fuel cell technology has emerged as a new reliable energy source that can replace fossil fuels, especially the polymer electrolyte membrane (PEM) fuel cell. PEM fuel cells are the most promising power source for various applications such as transportation due to its high efficiency, high power density, low operating temperature and long duration [1–3]. The most important component in a PEM fuel cell is the bipolar plate. The bipolar plate constitutes 60–80% of the total weight and 30–50% of the total cost for the fuel cell system [4–7]. Bipolar plate performance plays a significant role in the overall fuel cell performance. The bipolar plate performs three main functions: distribution of gases homogeneously over the entire area of the single cells, separation of the fuel and oxidant gases, prevention of leakage and collection of the current produced from the electrochemical reactions. However, the key function of the bipolar plate is to pass electrons based on its conductivity [8–10].

Early bipolar plates were made from metals such as stainless steel, aluminum and titanium. The major drawback for the metal based bipolar plate is corrosion. Despite having excellent electrical conductivity, mechanical strength and being easy to fabricate, metal based bipolar plates may induce the formation of oxide layers on the surface, leading to corrosion attacks and reducing the electrical performance of the bipolar plate [11]. This limiting factor associated with metallic bipolar plates has led to the development of graphite bipolar plates. Graphite possesses good electrical conductivity and excellent corrosion resistance. However, graphite also has weaknesses, such as its poor mechanical strength and the difficulties associated with fabrication. Bipolar plates made from polymer composites have been developed to replace metallic bipolar plates due to their better corrosion resistance, low cost, low mass and ease of manufacturing [11,12]. Polymer composites consist of a polymer matrix composed of a thermosetting or thermoplastic polymer with the addition of conductive fillers, such as graphite(G), carbon black (CB), carbon nanotubes (CNTs) and carbon fibers (CF). Carbon fibers are attractive because they have the ability to improve the thermal, electrical and mechanical properties of a composite [13–15]. CF is well known as a reinforcement agent

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due to its high strength and modulus that significantly contribute to a composite's performance. Composites containing CF are often stronger and are much lighter than steel [16]. The superior properties of CF make them good candidates for bipolar plate materials. Common CFs are produced by first converting the carbonaceous precursor into a fiber form and then cross-linking to render it infusible. Finally, the cross-linked precursor fiber is heated to temperatures ranging from 1200 to 3000 °C in an inert atmosphere to eliminate all of the non-carbon elements, finishing the conversion of the precursor into carbon fiber [17]. Depending on the precursors, carbon fibers can be divided into three types: PAN-based, pitch based and rayon based. The most common carbon fibers are produced from either polyacrylonitrile (PAN) or mesophase pitch precursors. PAN-based carbon fibers have excellent strength but a fairly low modulus. Additionally, the strength decreases as the modulus increases. Mesophase pitch-based fibers possess a lower tensile strength, but have the potential to develop modulus levels approaching the theoretical limit of graphite and have much better thermal and electrical conductivity than PAN-based fibers [18]. Previous studies showed that PAN types CF is suitable for the development of a composite bipolar plate [19,20].

Composite bipolar plates using CF was developed a years ago using fabric CF and short CF. The addition of CF provides good mechanical properties to the composite. However, excess CF increases the permeability of the composite [21]. Based on previous work, increasing the CF concentration also increases the resistivity, leading to poor electrical properties [19]. This phenomenon has been observed as a result of the orientation of the CF, which is perpendicular to the current flow. A fabric type CF in a layer form was developed to overcome this problem [22].

In this paper, the potential of milled carbon fiber (MCF) as secondary filler in graphite/epoxy composites is investigated. The investigation process includes the determination of the electrical and mechanical properties of the composite. This study also focuses on the potential of using the GEM model to predict the electrical properties of a conductive polymer composite with a high degree of filler loading.

The electrical properties of a composite are often described by the percolation theory for a low degree of filler content. The electrical conductivity of the composite is near that of the conductivity of the insulating material. When the filler content is greater than the critical content determined by the percolation threshold, the electrical conductivity rapidly increases [23]. From the percolation theory, McLachlan developed a model to predict the conductivity of a binary mixture with anisotropic grain structures. The equation described the electrical conductivity in terms of the volume fraction and the critical component factor [24]. The GEM model was developed by McLachlan due to the restriction of the Bruggeman effective media theory and the classical percolation theory [25]. The GEM model uses the same parameter as the classical percolation and may be used to predict the various compositions. As a result of studies carried out by previous researchers, the GEM model is able to effectively predict the electrical conductivity of a polymer composite consisting of single or multiple fillers [26,27]. The GEM model for a two component system is:

$$\frac{(1-\varnothing)\left(\sigma_l^{\frac{1}{t}}-\sigma_m^{\frac{1}{t}}\right)}{\sigma_l^{\frac{1}{t}}+A\sigma_m^{\frac{1}{t}}}+\frac{\varnothing\left(\sigma_h^{\frac{1}{t}}-\sigma_m^{\frac{1}{t}}\right)}{\sigma_h^{\frac{1}{t}}+A\sigma_m^{\frac{1}{t}}}=0 \tag{1}$$

$$A=\frac{1-\varnothing_c}{\varnothing_c} \tag{2}$$

where the electrical conductivity, σ_m is a function of the filler volume fraction Ø, t is an exponential component, A is related to the percolation threshold of a composite, and σ_l and σ_h are the electrical conductivities of the polymer and the filler. The exponential component, t is the critical exponent that may be adjusted to determine the morphology of the conductive particles. A universal value for t in a composite system with spherical particles is 1.65–2.0. The main advantage of the GEM model is that it can be modified to predict the electrical conductivity of multiple filler systems. In this paper, the GEM model is adapted to predict the electrical conductivity for a highly conductive composite.

Based on equation (1), parameter t has a significant effect on the outcome, which is the electrical conductivity of the composite. In this study, a new approach is used to determine t and predict the electrical conductivity of the composite. The parameter t can be estimated based on the individual t value for each filler. From this study, the t value for a single filler synthetic graphite is 2.41 for the in-plane conductivity and 2.51 for the through-plane conductivity. The t value for a single filler carbon fiber is 1.19 for the in-plane conductivity and 1.10 for the through-plane conductivity. For a multiple filler system, the t value may be estimated by taking the mean value as the combined t value. Considering this, a new method to determine t is defined by:

$$t=\frac{i}{n}\frac{(a+b+\ldots+i)}{n} \tag{3}$$

where a and b are the t values for the primary and secondary fillers and n is the total number of conductive fillers in the composite system. This equation is valid for both the in-plane and through-plane electrical conductivities. Using this method, the electrical conductivity of the composite may be predicted.

2. Experimental

The composite was prepared by melt compounding. Milled carbon fibers (MCF) were obtained from ShenZhenYataida High-Tech. Co., Ltd with a density of 1.75 g/cm^3, a diameter of 7 μm and an average length of 165 μm as reported by manufacturer. MCF has been chosen for its suitability of dispersion in epoxy matrix through mechanical mixing. Synthetic graphite (SG) was obtained from Asbury Carbon, New Jersey with a surface area of 1.5 m^2/g and a particle size of 74 μm. The matrix is an epoxy-based bisphenol-A with a viscosity of 6 Poise and procured from US Composites. The hardener agent is 4-aminophenylsulphone, which was procured from Fisher Scientific (M) Sdn. Bhd, Malaysia. A diamine hardener agent (tetra functional) is used to ease the rapid and compact cross-linking of the epoxy resin. Epoxy was chosen because of its unique properties such as good mechanical properties, low density and electrical resistance [23].

Epoxy resin and the curing agent were mixed at 3:1 ratio in a high speed mechanical mixer, model RM 20-KIKA-WERK, at 1200 rpm for 40 s. Then, the conductive filler was added to the mixer and mixed for another 5 min at room temperature. After the mixing was completed, the composite mixture was poured into a steel mold at150 °C and cured at a molding pressure of 1800 psi for 90 min.

The in-plane electrical conductivity of the composite is measured with a four point probe technique using a Jandel four point probe and an RM3 test unit. The through-plane electrical conductivity of the composite was measured with a through plane electrical conductivity tester manufactured by ZBT in Duisburg, Germany [28]. The structure of the composite was observed using a scanning electron microscope Model Zeiss MA10. The mechanical properties of the composite were determined based on the ASTM