

Optimization of a Carbon Composite Bipolar Plate for PEM Fuel Cells

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ABSTRACT

A carbon composite bipolar plate for PEM fuel cells has been developed that has high electrical conductivity, high strength, light weight, is impermeable, and has the potential for being produced at low cost. The plate is produced by slurry molding short carbon fibers into preform structures, molding features into the green body, and using chemical vapor infiltration to strengthen the material, give it high conductivity, and densify the surface to make it impermeable. Current efforts have focused on optimizing the fabrication process and characterizing prototypical components.

INTRODUCTION

The significant and growing interest in fuel cells for stationary power and transportation applications has been demonstrated by the attention these technologies are receiving from both government and industry, and particularly from the automotive sector [1]. Interest for vehicular applications has focused on the proton exchange membrane fuel cell (PEMFC) because of its low-temperature operation and thus rapid start-up. Currently, challenges for PEMFC technology for automobiles include reducing the cost and weight of the fuel cell stack, the goal being a ~50 kW system of <\$40/kW and <133 kg in mass. One of the key components is the bipolar plate, which is the electrode plate that separates individual cells in a stack [2]. The reference design requires the bipolar plate to be high-density graphite with machined flow channels. Both material and machining costs are prohibitive (\$100-200/plate), and this has led to substantial development efforts to replace graphite. The bipolar plate requirements include low-cost materials and processing (goal of <\$10/kW for the component), light weight, thin (<3mm), sufficient mechanical integrity, high surface and bulk electronic conductivity, low permeability (boundary between fuel and oxidant), and corrosion resistance in the moist atmosphere of the cell (<16 $\mu\text{A}/\text{cm}^2$) [3].

The bipolar plate approach developed at Oak Ridge National Laboratory (ORNL) uses a low-cost, slurry-molding process to produce a carbon-fiber preform. The molded, carbon-fiber component could have an inherent volume for diffusing fuel or air to the electrolyte surface or impressed, flow-field channels. The bipolar plate is made hermetic through chemical vapor infiltration (CVI) with carbon. The infiltrated carbon also serves to make the component highly conductive. The technique has the potential for low-cost, large-scale production as the material costs are low, the fiber preforms can be produced in continuous processes similar to felt or paper production, and the infiltration can be accomplished in very large-scale batch or possibly continuous processes.

This paper reviews current work in characterizing the carbon composite materials with differing loadings of carbon particulate filler. The filler was added in an attempt to reduce the need for fiber material and make surface sealing easier. Surface roughness was characterized using profilometry and infrared imaging was used to identify flaws in sample coupons. In-plane shear stress was measured to gain an understanding of the materials' behavior under torsion, which may occur during stack assembly. Fracture toughness was measured so that when combined with strength data, previously determined, it could aid in determining the size of cracks that the material can tolerate.

EXPERIMENTAL

Slurry-Molding

Fibrous component preforms for a sub-scale plate (120 x 140 x 2.5 mm) are prepared by a slurry molding technique using 10 μm -diameter, 100- μm -long carbon fibers (Fortafil 3(c) 00 PAN-based, Fortafil Fibers, Inc., Rockwood, TN) suspended in water containing phenolic DUREZ[®] resin (Occidental Chemical Corp., Dallas, TX) [4]. For some samples, a graphite particulate filler, sieved to $\sim 7 \mu\text{m}$, was added to the fiber at 15 and 30 mass % (Asbury M850 graphite, Asbury Carbons, Inc., Asbury, NJ). The fiber-to-phenolic mass ratio is 4:3. A vacuum-molding process produces an $\sim 18 \text{ vol } \%$ fiber, isotropic preform material containing particles of phenolic. After drying, a set of brass molds are used to impress features, such as gas channels, into the preform material at 150°C and a pressure of 10 kPa. The phenolic binder serves to provide green strength and geometric stability after curing in the mold.

Chemical Vapor Infiltration.

The surface of the preform is sealed using a CVI technique in which carbon is deposited on the near-surface material in sufficient quantity to make it hermetic. The depth of infiltration is governed by the inherent competition between the kinetics of the surface reactions that produce the deposited material and the mass transport mechanism that allows the reactants to diffuse to the internal volume of the material [5]. The result is that the more rapid the kinetics of deposition as compared to mass transport, the more likely material will be deposited near the surface and the reactants will not significantly penetrate into the thickness of the porous preform. In a review of carbon deposition, Delhaes [6] describes depth penetration of CVI carbon as a function of temperature. The higher the temperature the more rapid the surface deposition kinetics, thus the smaller penetration depth allows the surface to be sealed and the bulk volume of the preform to retain a large volume fraction of porosity.

A high surface-to-volume CVI reactor was used to minimize soot formation and allow the efficient infiltration of the fibrous preform. Based on Delhaes [6] and Bammidipati, et al. [7], an infiltration temperature of 1500°C was selected with methane (chemically pure, Air Liquide, Houston, TX) as the precursor. Reduced pressure (8 kPa) also suppresses the formation of soot. Flow rates of $1000 \text{ cm}^3/\text{min}$ methane in $2500 \text{ cm}^3/\text{min}$ argon are used. A processing time on the order of 4 h was determined to be necessary to obtain sealed surfaces for typical components without particulate filler. In

addition, during the high temperature CVI processing the phenolic present in the preform is pyrolyzed. Further details can be found in Besmann, et al. [8].

Characterization

The surface roughness of the bipolar plates was measured using a Rodenstock RM600 laser profilometer with version 3.27 software (Rodenstock GmbH, München, Germany).

Infrared (IR) images to identify flaws/delamination in bipolar plate material samples were obtained using a high-resolution IR camera (Radiance 1t, Amber/Raytheon, Goleta, CA) with a 25 mm lens. The camera contained a 256 x 256 pixel indium antimonide sensor that was sensitive in the 3-5 μm waveband. Each pixel represented 100 μm x 100 μm projected area of the bipolar plate and had a temperature resolution of $\pm 0.1^\circ\text{C}$ throughout the examined temperature range. The camera has a high speed (12 bit) video bus that was used to digitally capture the temperature distribution field. The bipolar plate specimens were imaged while they were on top of a hot plate that induced a temperature gradient through their thickness. Differences in surface temperature, which are recorded with the IR camera, are indicative of inhomogeneities in the thermal conductivity of the material (e.g.- delaminations, defects). Figure 1 illustrates the configuration of the thermal imaging system.

In-plane shear strength tests were carried out per ASTM C1292. Because the test specimens were smaller than those recommended in C1292, stainless-steel end tabs were adhesively bonded to the specimens. Both the specimens and the V-notches were machined using a diamond wheel. The tests were carried out at a constant cross-head displacement rate of 0.05 mm/s using an electromechanical testing machine and an Iosipescu fixture (Fig. 2).

Fracture toughness measurements were determined by a double torsion test. The geometry of the test specimen consists of a plate which is notched to form two beams. Each of these beams is loaded in torsion by four-point bending at the ends, causing the crack to propagate through the center of the specimen (Fig. 3) [9]. In this specimen configuration the crack profile is not straight through the thickness, but extends further along the tensile side of the plate to form a curved crack front. Both the specimens and the notch were machined using a numerically-controlled grinder equipped with a thin diamond wheel. The tests were carried out at ambient conditions using an electromechanical testing machine at a constant cross-head displacement rate of 0.1 mm/s using an in-house developed fixture. To determine the compliance of the load train and specimen, test specimens with different notch lengths were evaluated.

RESULTS

An optical image of a cross-section of a bipolar plate sample can be seen in Fig. 4. The surface is sealed with a layer of carbon, yet significant porosity is visible in the interior of the material. The average density of infiltrated bipolar plates is $\sim 1.2 \text{ g/cm}^3$ (theoretical density of carbon is 2.26 g/cm^3). Figure 4 also contains representative surface profile measurements of a prototypical

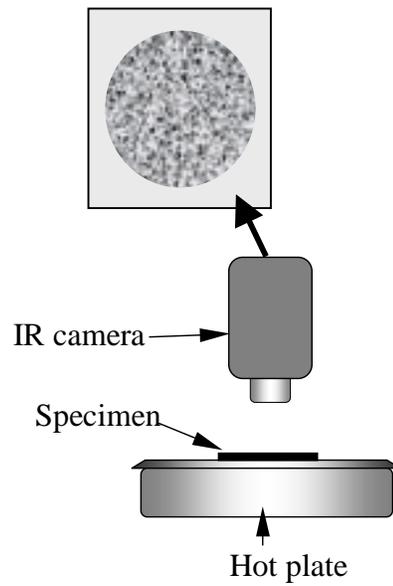


Figure 1. Infrared imaging system used for determining delamination and defects in bipolar plate samples.

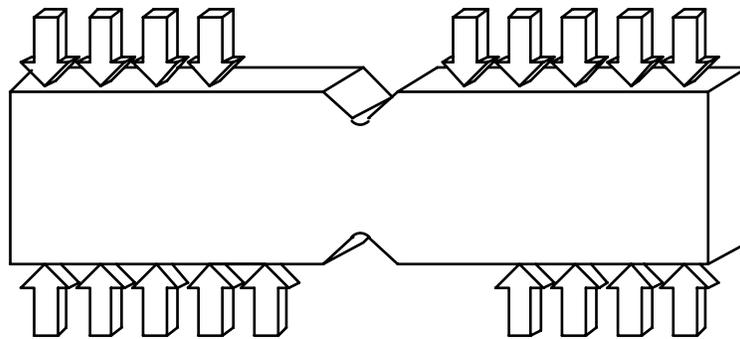


Figure 2. Iosipescu loading configuration for applying asymmetric bending to determine in-plane shear strength.

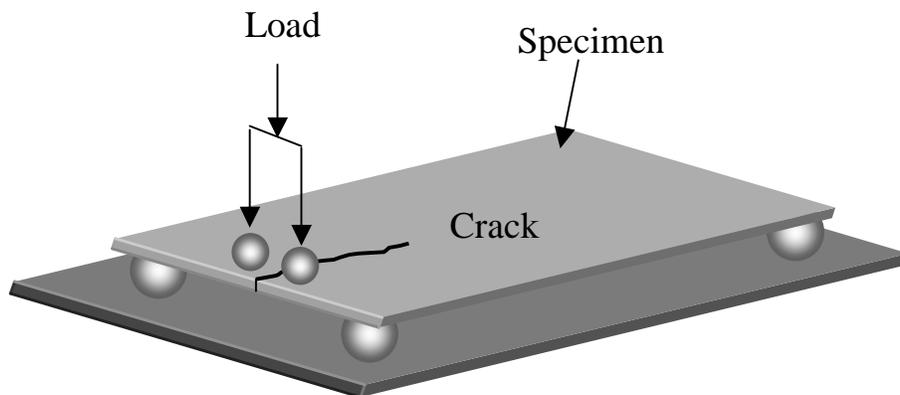


Figure 3. Double-torsion test method for determination of fracture toughness.

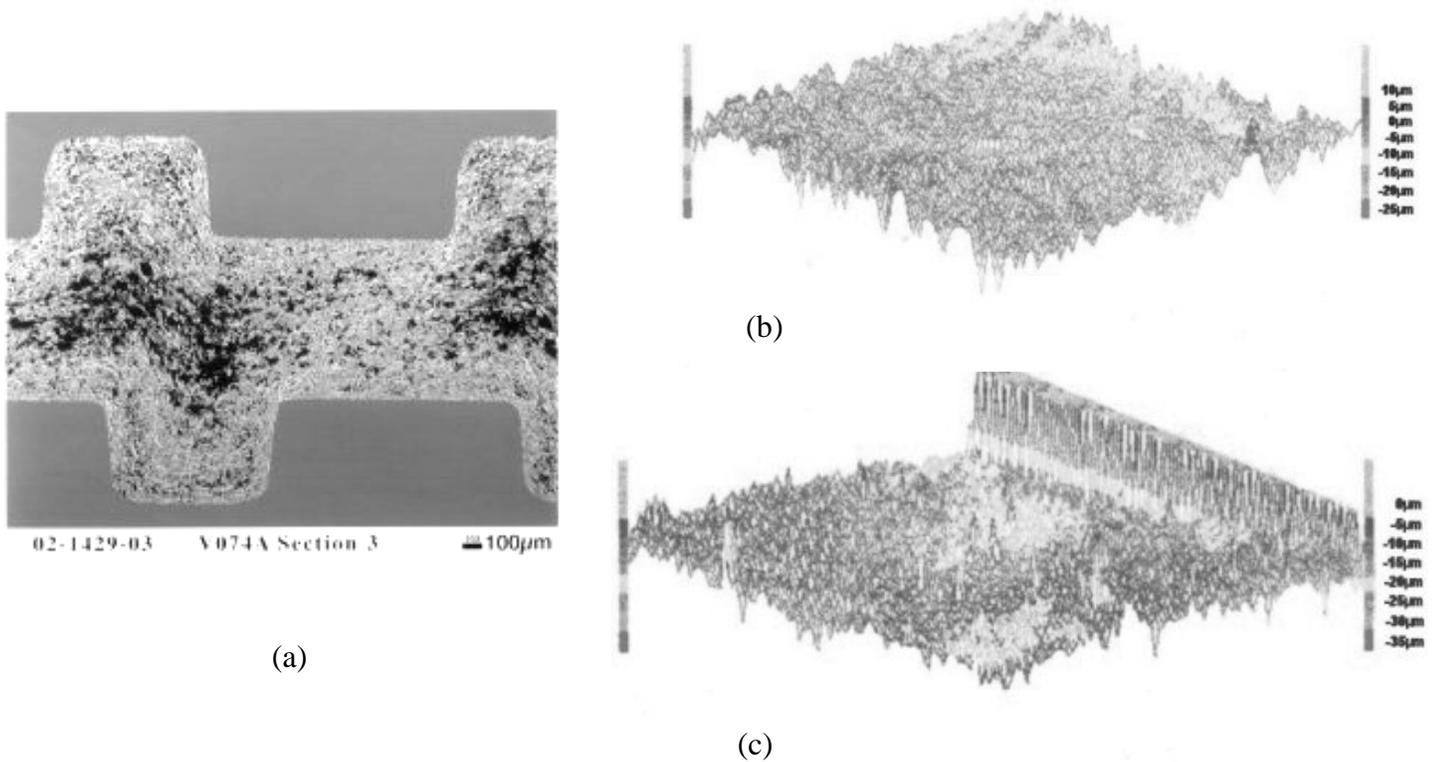


Figure 4. Prototypical carbon composite bipolar plate: (a) optical image of a polished cross-section, (b) surface profile of top flat, and (c) surface profile of bottom of a channel and its sidewall.

bipolar plate. Measurements indicate rms surface roughness values that vary between 2.5 and 4.5 µm. It is also apparent that in certain areas, such as in the corner of a sidewall of a channel, there is a higher degree of roughness, with depth variations of ~30 µm.

The integrity of bipolar plate material was assessed using thermal imaging. Figure 5 shows a series of IR images taken of 50x50x2 mm samples. These were cut from a larger plate after slurry-molding and curing, but before CVI. After CVI to seal the surface of the plates they were examined using the thermal imaging system. Gray-scale changes indicate delamination, with all but one sample indicating that they are intact. There does appear to be some damage around the edges of the samples, most likely due to local delamination due to cutting of the larger plate into the smaller samples.

Table I gives the results of the shear stress measurements using the Iosipescu method. An example of a stress-displacement curve obtained for the material is shown in Fig. 6. These values are indicative of a relatively torsion resistant material, particularly given the low density of the carbon composite. There appears to be little effect of filler content within the concentration utilized. The stress-displacement curves indicate little delamination or other failures until ultimate failure. Table II lists the results of the fracture toughness measurements, the values of which are fairly low, again with little effect of filler content.

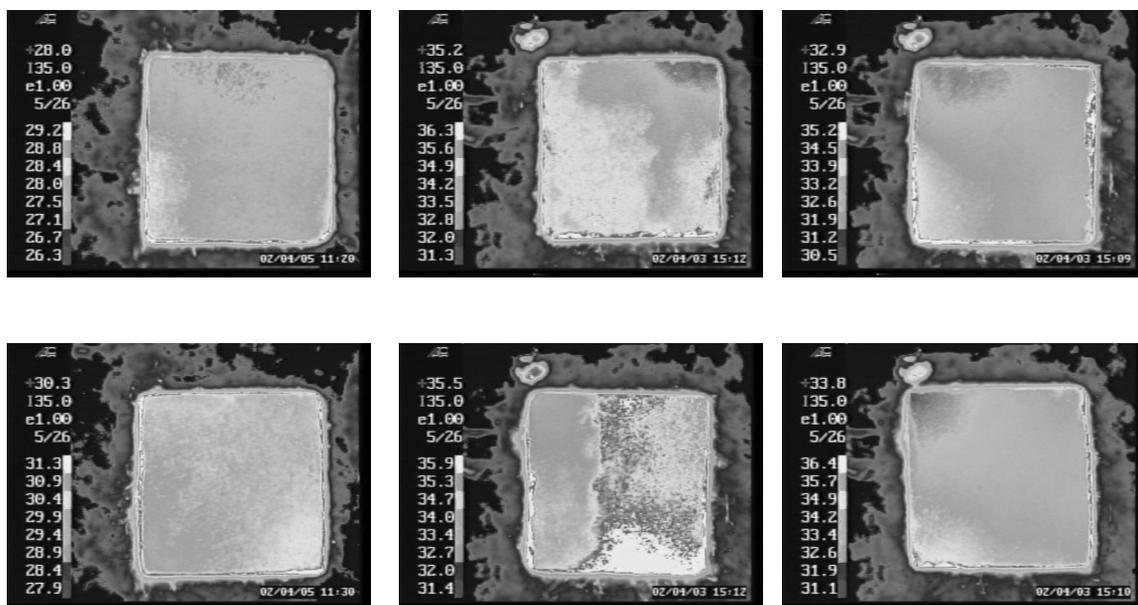


Figure 5. Infrared images of carbon composite bipolar plate material samples revealing delamination and edge defects.

Table I. Shear stress measurements shows little correlation with percent filler.

<u>Sample (% Filler)</u>	<u>Strength Meas.</u>
PMP10R (0%)	25.9 ± 9.9 MPa
PMP10T (0%)	19.3 MPa (1 test)
PMP09K (15%)	24.4 ± 11.8 MPa
PMP11E (30%)	43.3 ± 2.7 MPa
PMP11G (30%)	17.1 ± 1.1 MPa
PMP11H (30%)	18.7 ± 4.7 MPa

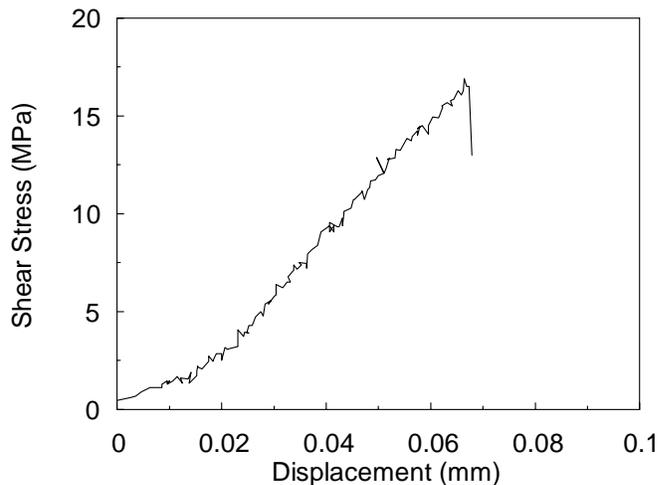


Figure 6. Typical stress-displacement curve for the shear test of the carbon composite bipolar plate material.

Table II. Fracture toughness of carbon composite bipolar plate material for different filler contents (assuming a Poisson's ratio of 0.2).

Sample (% filler)	Fracture Toughness (K_{IC} , $\text{MPa}\cdot\text{m}^{1/2}$)
PM10 (0)	2.55±0.19
PM09 (15)	3.24±0.11
PM11 (30)	2.84±0.36

DISCUSSION

Surface roughness can be an important issue for bipolar plate materials. The plate must contact the electrolyte membrane, as well as any intervening layers that are often used to support catalyst material. Thus, high surface roughness can affect electrical contact between mating surfaces. The relatively modest surface roughness of much of the bipolar plate areas is within the bounds of acceptability, as indicated by relatively good electrical behavior previously reported [8]. Surface roughness can also affect water management within a PEMFC, causing holdup of water and thus blocking of channels, particularly if wetting is a problem. This issue will need further exploration.

The results of the thermal imaging measurements, which were a first attempt to use this technique in this application, are encouraging. It is expected that in development and production there will be a need for a rapid and definitive technique for non-destructive evaluation of plates in production. Although it is likely that plates with some delamination or other flaws will still meet acceptability criteria, thermal imaging offers at least the opportunity to develop a baseline screening system.

As fuel cell stacks are assembled it is expected that relatively high pressures will be needed to obtain good electrical contact and aid in sealing the edges against leakage. Given that upwards of 100 plates will be needed in a stack, and that it is likely that pressure will be applied via techniques such as posts on each corner running the length of the stack, it is important that cell components be able to withstand significant tension and torsion, as well as compression. The Iosipescu test methodology was therefore viewed as particularly applicable to assess the resistance of the bipolar plates to torsional loading. The relatively low fracture toughness of the material is a concern for handling and assembly of the plates, although the implications are uncertain due to the highly inhomogeneous and anisotropic nature of the components.

CONCLUSIONS

The technique of slurry-molding carbon fibers followed by chemical vapor infiltration with carbon has resulted in relatively strong and light-weight PEMFC bipolar plates with good electrical and corrosion properties, and the potential for low cost manufacturing. Examination of the surface roughness of molded and infiltrated plates revealed relatively smooth surfaces that will facilitate sealing and electrical contact. Thermal imaging has been demonstrated as a potential tool for non-destructive evaluation of bipolar plate components, and has revealed that as-processed materials does not delaminate. Mechanical property measurements of the shear stress resistance of the

carbon composite bipolar plate material are encouraging, with relatively good strengths that are likely to withstand assembly and use of the plates. Fracture toughness values are low, but may be misleading due to the inhomogeneity of the material.

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