

Development of bipolar plates for fuel cells from graphite filled wet-lay material and a compatible thermoplastic laminate skin layer

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Abstract

In this paper a method with the potential to lead to the rapid production of thermoplastic polymer composite bipolar plates with improved mechanical properties, formability, and half-cell resistance is described. In our previous work it was reported that laminate structure composite bipolar plates made with a polyphenylene sulfide (PPS) based wet-lay material as the core and a polyvinylidene fluoride (PVDF)/graphite mixture as the laminate exhibited improved formability, through-plane conductivity, and half-cell resistance over that of wet-lay based bipolar plates. However, the mechanical strength of the laminate plates needed improvement. In this work laminate polymer composite plates consisting of a PPS/graphite-based laminate mixture and a PPS based wet-lay core are manufactured in an effort to improve mechanical strength. Additionally, our existing channel design has been altered to reduce the channel depth from 0.8 to 0.5 mm in an effort to improve the half-cell resistance by reducing the total plate thickness. The plates are characterized by their half-cell resistance and mechanical properties at ambient and elevated temperatures. The PPS based laminate plates exhibited half-cell resistances as low as $0.018 \Omega \text{ cm}^2$, tensile strength of up to 37 MPa, and flexural strength of up to 60 MPa at ambient temperature. The laminate bipolar plates can be manufactured in several ways with two of them being discussed in detail in the paper.

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1. Introduction

Bipolar plates are by weight, volume, and cost one of the most significant parts of a fuel cell stack. Bipolar plates must exhibit excellent electrical conductivity, adequate mechanical stability, corrosion resistance (for metal plates/metal coated fillers), and low gas permeability. Furthermore, bipolar plates must be amenable to a rapid, low cost processing methodology if they are to be widely used in the automotive industry.

The endeavor to develop a suitable material for bipolar plate production is based on the following requirements:

- electrical conductivity ($>100 \text{ S/cm}^a$);
- low permeability ($<2 \times 10^{-6} \text{ cm}^3 (\text{cm}^2 \text{ s})^{-1})^a$;
- corrosion resistant ($<16 \mu\text{A cm}^{-2})^a$;
- tensile strength ($>41 \text{ MPa}^b$);
- flexural strength ($>59 \text{ MPa}^b$);
- high thermal conductivity ($>10 \text{ W (m K)}^{-1})^b$;
- chemical and electrochemical stability;
- low thermal expansion;
- efficient processability.

The electrical conductivity target is specified as bulk, or in-plane conductivity. Although no target has been specified for through-plane conductivity, it is thought to be an important measurement because the through-plane direction is the route that electrons travel in an operating fuel cell. Corrosion resistance is mainly a

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^a Department of Energy (DOE) and FreedomCAR specified targets [1].

^b Plug Power targets [2].

concern with metal plates and metal coated fibers, but it can also apply to polymer systems that may dissolve electrochemically in a fuel cell. The mechanical targets are specified for ambient temperature, while targets at elevated temperatures have not yet been specified. Efficient processability incorporates several factors, including material and production costs and the ability for rapid, continuous manufacturing.

Traditional materials for producing bipolar plates are sintered graphite and metal. Graphite plates have been used because they possess high electrical conductivity, corrosion resistance, and a lower density than that of metals [3]. Problems with graphite plates include brittleness and the cost associated with machining channels into the surface. Because the material and machining cost of graphite plates are exorbitant, at least for the automotive industry, the development of a more suitable material has drawn extensive research. Metal bipolar plates such as stainless steel, aluminum, and titanium have gained attention for their high electrical conductivity, mechanical strength, and negligible gas permeability [3–5]. The main advantage with metal plates is that channels can be embossed or etched into the surface, eliminating or reducing considerable time and cost associated with machining. However, because metals are subject to corrosion and leaching of ions, a coating must be applied to the surface to provide a protective layer. Unfortunately, the protective coating is highly electrically insulating and drastically increases the interfacial resistance of the metal bipolar plates. Many materials have been investigated to coat metal plates such as polypyrrole [6] and nitrides [7,8], but it has been shown to be difficult to apply a protective layer thin enough to maintain low interfacial resistance while avoiding the existence of pinholes or defects in the surface, ultimately leading to bipolar plate failure.

In an effort to improve on sintered graphite, researchers at Oak Ridge National Laboratory (ORNL) have developed carbon/carbon composite bipolar plates by slurry molding carbon fiber and phenolic resin [9]. ORNL researchers claim that the plates have high electrical conductivity and adequate mechanical stability. However, the manufacturing step in the production of carbon/carbon plates is complex and expensive due to a chemical vapor infiltration (CVI) process. CVI operates at very high temperatures for extended times, sealing the carbon/phenolic resin preforms by depositing carbon onto the surface. Unfortunately, channels must still be machined into the surfaces of the carbon/carbon plates.

Graphite-based polymer composite bipolar plates have shown to potentially meet target requirements. They offer advantages such as lower cost, higher flexibility, and are lighter in weight when compared to metallic and graphite plates. The gas flow channels can be molded directly into the surfaces of the plate, eliminating the need for a costly machining step. Researchers at Los Alamos National Laboratory (LANL) have developed a bipolar plate based on a vinyl ester thermosetting polymer resin and graphite powder [2,10]. Bulk conductivities were measured to be as high as 85 S/cm, with tensile and flexural strength at 25 and 38 MPa, respectively, which are still lower than the specified targets. A compression molding process was used for producing the bipolar plates in which channels were formed in less than 10 min [11,12]. The advantage in using a thermosetting polymer

is that when the polymer is heated and compression molded, the plate cures and does not require subsequent cooling. Therefore, the plate can be immediately released from the mold. However, a postcure may be necessary, and can take as long as 1 h to complete [11]. Blunk et al. [13] have developed an epoxy-based system using expandable graphite that has exhibited low resistances. However, the flexural strength of the plates did not reach the target value. Although the graphite filled epoxy system has shown potential for use in bipolar plate production, the generation of the epoxy bipolar plates requires a 20 min curing time and the expandable graphite is anticipated to be expensive to manufacture. The epoxy-based bipolar plates also require machining of channels. Polyvinylidene fluoride (PVDF), a thermoplastic fluoropolymer matrix, has been used with graphite particles and carbon fiber to produce bipolar plates [14]. Bulk conductivity values have reached 109 S/cm, exceeding the DOE target. However, the flexural strength was only 42.7 MPa, lower than the goal of 59 MPa. Liquid crystalline polymer/graphite mixtures have been considered for bipolar plate production because of their ability to be injection molded due to the low viscosity of the polymer [15]. The injection molding process allows for a relatively short cycle time of 30 s. Bulk conductivities have reached values as high as 100 S/cm, just reaching the minimum target, but no mechanical properties or through-plane conductivities were reported, and it is anticipated that the mechanical properties may not reach the targets.

Huang and co-workers [16] have reported the development of conductive polymer composite materials generated by means of a wet-lay process. The wet-lay materials were compression molded to form highly conductive and strong bipolar plates. Bulk conductivities (in-plane) and mechanical properties of the plates were higher than that of any other polymer composite plates (see Table 1). However, the through-plane conductivity, half-cell resistance, and formability of the plates needed further improvement.

Cunningham and co-workers [16–18] have improved on the through-plane conductivity, half-cell resistance, and formability of the wet-lay based bipolar plates by using a laminate approach. A PVDF/graphite mixture in powder form was applied to the surfaces of a polyphenylene sulfide (PPS) based wet-lay core material. The PVDF based laminate plates exhibited bulk conductivities greater than 300 S/cm, well above the DOE target. Compared to the bipolar plates consisting of wet-lay material only, the laminate bipolar plates exhibited an increase in through-plane conductivity of 25–35%, as well as a decrease in half-cell resistance by a factor of up to 5. The tensile and flexural strengths were measured to be 34 and 54 MPa, respectively, both falling short of the target value but higher than those of any other polymer composite material used for the production of bipolar plates.

In this paper a PPS based laminate structure is investigated as a way to improve the mechanical properties over those of PVDF based laminate plates while maintaining sufficient formability and half-cell resistance of the bipolar plates. The laminate bipolar plates are generated by two methods. One is to put laminate mixture (in powder form) on the surfaces of the wet-lay sheet stacks and compression mold at a temperature to melt

Table 1
Comparison of conductive and mechanical properties for several polymer composite materials used in the production of bipolar plates

Source	Polymer	Graphite/glass or carbon reinforcing fiber (wt/wt%)	Conductivity (S/cm)		Mechanical strength (MPa)	
			In-plane	Through-plane	Tensile	Flexural
Target			>100 ^a	–	41.0 ^b	59.0 ^b
GE [14]	PVDF	64/16 CF	109	–	–	42.7
LANL [11]	Vinyl ester	68/0	60	–	23.4	29.6
Plug Power [2]	Vinyl ester	68/0	55	20	26.2	40.0
DuPont [22]	–	–	–	25–33	25.1	53.1
Virginia Tech [16]	PET	65/7 GF	230	18–25	36.5	53.0
Virginia Tech	PPS	70/6 CF	271	19	57.5	95.8
Virginia Tech [19]	15% PVDF laminate	70/6 CF	350	14–32	32.7	54.4
Virginia Tech	20% PVDF laminate	80/6 CF	251	15–26	25.0	30.6

^a DOE target for conductivity [1].

^b Plug Power targets for mechanical strength [2].

the polymer which is called the one-step molding method. The other is to generate a consolidated wet-lay composite plaque first and then add composite powders to the surfaces of the flat plaque followed by compression molding the composite to impart the channels. This is called the two-step molding method. We will assess the properties of the bipolar plates made by the two different methods and discuss the advantages and disadvantages of the processes. The evaluation will be focused on the mechanical properties of the composites, as well as the half-cell resistance and surface appearance of the bipolar plates.

2. Experimental

2.1. Materials

For generating the wet-lay material, a thermoplastic polymer used in the form of fibers was PPS (6 mm long) at levels in the range of 13–40 wt%. Conoco carbon fibers (nominal length 1/2 in.) were used as reinforcing fibers in the range of 6–9 wt%. Timrex[®] KS-150 and Timcal[™] TC-300 graphite particles were used as the conductive filler in the range of 50–80 wt%. Microglass (1 wt%) was also added to help particle retention in the wet-lay preforms. PVDF (Kynar 761 from Arkema) was used with TC-300 graphite particles as the first type of laminate mixture. PPS powder (Primef[®] from Solvay) was used with TC-300 graphite particles as the second type of laminate mixture.

2.2. Production and molding of wet-lay composite sheet material

In Fig. 1 is shown the scheme for production of the wet-lay composite materials and bipolar plates. Wet-lay sheets containing a thermoplastic fiber, reinforcing fiber, conductive graphite particles, and microglass were generated with a slurry-making process on a Herty papermaking machine donated by DuPont. The slurry contained 1 wt% solids in water. First, the cut thermoplastic fibers were added to the water in a pulper and agitated for 10 min. Next, the cut reinforcing fibers, graphite particles, and microglass were added and mixed for 3–6 min. The slurry was then pumped into a head box containing a sieve screen, generating a sheet of porous material. The sheet was continuously rolled through an oven at the melting point of the polymer to continue drying and partially melt the thermoplastic fibers.

The procedure carried out by Huang and co-workers [16] for making a test sample or bipolar plate from wet-lay material was followed in this work. The material was cut to fit the shape of a mold and stacked to provide a desired thickness. The mold was then placed between platen heaters of a hydraulic press set at approximately 20 °C above the melting point of the thermoplastic fiber (300 °C for PPS based composites). The material was compression molded for 7–10 min at a constant applied pressure of 1000 psi. The platen heaters were turned off and the mold was allowed to cool to 30 °C while still under pressure. The mold was then removed from the press and the flat plaque or bipolar plate was taken out of the mold. Although a batch process

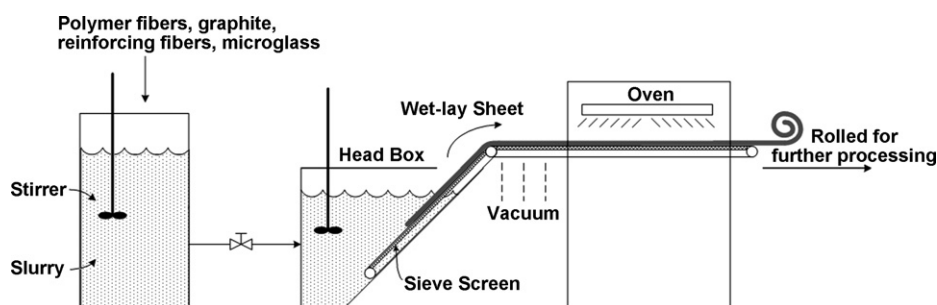


Fig. 1. The process to develop wet-lay polymer composite sheets.

was used in this work, a continuous process is envisioned in the future.

2.3. Novel laminate approach

In this paper two methods to produce bipolar plates are described. The first, called the one-step method, consisted of initially covering the bottom surface of the mold with a desired amount of laminate mixture and spreading it evenly over the surface. Next, a pre-determined number of wet-lay sheets were stacked on top of the laminate mixture. Finally, another layer of laminate (same amount as placed in the bottom of the mold) was spread evenly on top of the wet-lay sheets. The mold was closed and placed into a heated press set at 300 °C for compression molding. Upon compression molding, the laminate powder penetrated into the porous wet-lay material (>50% voids) to help construct a well bonded polymer network throughout the entire plate. The second process is called the two-step method. First, the wet-lay sheets by themselves were placed into a heated mold and compression molded to form a pre-consolidated flat plaque. After being removed from the mold, laminate powder was added to the bottom surface of the mold and on top of the flat plaque similar to the one-step method. The mold was closed and placed into a heated press for compression molding. The molding temperature for this method was set at approximately 20 °C above the melting point of the polymer in the laminate mixture (200 °C for PVDF, 300 °C for PPS). In the two-step method, the sole form of bonding between the skin layer and the wet-lay core was adhesion between the surfaces. The level of laminate powder added to the surfaces of the wet-lay material was varied from 15 to 25 vol% (on each side) to evaluate how differences in the level of laminate layer would affect the composite properties. Two channel designs of 0.8 mm wide × 0.8 mm deep and 0.8 mm wide × 0.5 mm deep were used to evaluate the dependency of half-cell resistance on channel depth of plates of similar

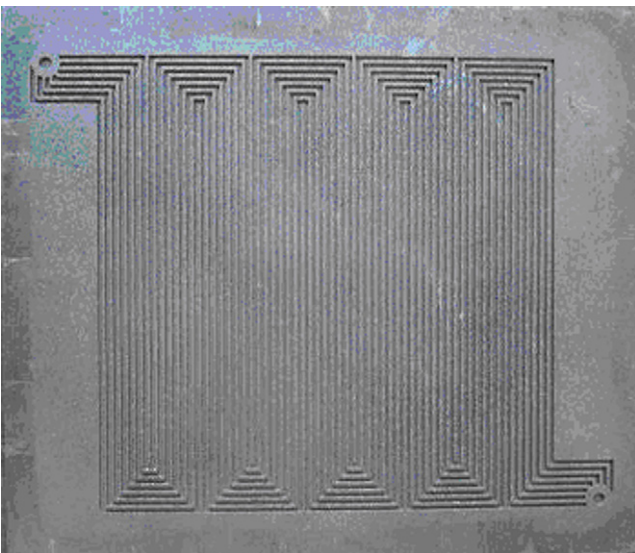


Fig. 2. One side of a PPS based laminate polymer composite bipolar plate showing the seven-channel parallel serpentine style design. Plate dimensions are 140 mm × 120 mm. Channel dimensions are 0.8 mm × 0.8 mm.

compositions. In Fig. 2 is shown a picture of a compression molded laminate bipolar plate of dimensions 120 mm × 140 mm with 0.8 mm × 0.5 mm and a parallel serpentine style channel design.

2.4. Measurement of mechanical properties

Tensile and flexural tests were performed on an Instron 4204 in accordance with ASTM D638 and D790 standards, respectively. Flexural tests were also performed on the PPS based laminates at 80, 100, and 120 °C to evaluate the strength in an environment that more accurately reflects that found in PEM fuel cells. The test specimen size was approximately 76.2 mm × 8.5 mm × 3.0 mm ($L \times W \times T$) and was cut from a flat plaque.

2.5. Measurement of in-plane and through-plane conductivity

In-plane (bulk) conductivity was measured according to ASTM Standard F76–86. Current contacts were placed at the four corners of the plaque allowing for a constant current to pass through the specimen. The voltage drop was measured across the specimen with a Keithley 2000 digital multi-meter at ambient conditions. Two characteristic resistances, R_A and R_B were measured. The plaque resistance, R_S , is obtained by solving the Van der Pauw equation:

$$\exp\left(\frac{-\pi R_A}{R_S}\right) + \exp\left(\frac{-\pi R_B}{R_S}\right) = 1 \quad (1)$$

The resistivity, ρ , is given by $\rho = R_S d$, where d is the thickness of specimen. The volume conductivity, σ_B , is defined as $1/\rho$.

Through-plane conductivities were measured based on a method proposed by Landis and Tucker [20]. A 76.2 mm × 76.2 mm plaque was placed between gold plated copper electrodes. Between the electrodes and sample was placed a piece of carbon paper (Toray TGP-H-120) to improve electrical contact between the electrodes and sample. The system was placed under an applied pressure of approximately 220 psi and the resistance was measured. The sample was removed, and the resistance of the test cell (including carbon paper) was measured again under the same conditions to obtain a “baseline” resistance. The sample resistance could then be calculated by subtracting the baseline resistance from the total resistance. The resistivity of the material was calculated by:

$$\rho = \frac{(R_T - R_{\text{baseline}})A}{L} \quad (2)$$

where ρ is the resistivity, A the cross-sectional area of sample, L the thickness of sample, and R_T and R_{baseline} are the total resistance and baseline resistance, respectively. The through-plane conductivity, σ_T was then calculated as $1/\rho$.

2.6. Measurement of half-cell resistance

To measure the half-cell resistance of a bipolar plate, an apparatus was set up similarly to the one used to measure

Table 2
Comparison of in-plane and through-plane conductivities for PPS and PVDF based laminate composites generated by the one-step and two-step methods

Material designating the amount and type of graphite contained in the wet-lay core	Processing method	In-plane conductivity (S/cm)	Through-plane conductivity (S/cm)
20 vol% PPS laminate with 70% TC-300 graphite in core	One-step	209–325	13–30
20 vol% PPS laminate with 70% TC-300 graphite in core	Two-step	164–226	13–29
20 vol% PVDF laminate with 70% TC-300 graphite in core	One-step	181–292	17–32
20 vol% PVDF laminate with 70% TC-300 graphite in core	Two-step	203–350	14–24

through-plane conductivity. A bipolar plate having length and width dimensions of 120 mm × 140 mm and an active area of 100 cm² was placed between the gold plated copper electrodes. Carbon paper (Toray TGP-H-120) was placed on both sides of the bipolar plate, and hence, in between the sample and electrodes. The size of the carbon paper was 100 mm × 100 mm in order to completely cover the active area. The sample was placed under an applied pressure of approximately 220 psi while a constant current of 250 mA was passed through the sample. It was acknowledged that in an operating fuel cell stack, the bipolar plates will be placed under 250 psi of pressure. However, due to the limitations of our apparatus, 220 psi was the maximum pressure applied to the bipolar plates. The potential was measured between the collectors and the half-cell resistance was calculated based on Ohm's law. The bipolar plate was removed, and the potential across the electrodes and carbon paper was measured to produce a baseline resistance. The baseline resistance, that is the resistance of the testing circuit excluding the bipolar plate but including carbon papers and electrodes, was measured after testing of the plate. This was done to ensure the stability of the baseline of the instrument and to evaluate the contribution of the bipolar plate to the total half-cell resistance.

2.7. Optical measurement of surface roughness

Surface roughness of bipolar plates was quantitatively measured using a TOPO 2-D Topography Analysis machine. The optical sensor measured the roughness along a 2 mm span of a channel that was compression molded into the surface. The optical sensor detected changes in the topography of the channel section in terms of peaks and valleys that emerged from a calculated centerline.

3. Results and discussion

3.1. Production of laminate composite materials and bipolar plates

It was found that the addition of a PVDF/graphite skin layer that covers PPS based wet-lay material improved through-plane conductivity, half-cell resistance, and the formability of channels into the surfaces [19]. However, the mechanical strengths of the PVDF based laminate plates did not quite meet the target values. Furthermore, there were concerns about compatibility and hence, adhesion of PVDF to the PPS based wet-lay. Therefore, it was believed that using PPS instead of PVDF in the

laminate powder mixture could provide a plate with sufficient strength due to the matching polymer structure and crystallization temperature range. All laminate bipolar plates or flat plaques discussed in this paper contained a PPS based wet-lay core with 70 wt% TC-300 graphite particles and 6 wt% carbon fibers.

3.2. Electrical conductivity of laminate composite materials

Electrical conductivity is one of the most important properties of bipolar plates. Huang and co-workers [16] have reported the electrical properties of wet-lay based composites compared to other polymer composite materials (see Table 1). PPS based wet-lay composites have exhibited in-plane (bulk) conductivities higher than 200 S/cm, well exceeding the DOE target of 100 S/cm. These values were also higher than any other polymer composite material with similar graphite loadings. PVDF based laminate composites have exhibited bulk conductivity values above 200 S/cm and improved through-plane conductivity values when compared to those of the wet-lay based composites [21]. PPS based laminate composites have also exhibited excellent in-plane conductivity values and through-plane conductivity values comparable to those of the PVDF based laminate composites as shown in Table 2. When materials of similar compositions differing only by processing method are compared, the laminate plates produced by the one-step method exhibit slightly higher conductivities, especially in the through-plane direction. This may be expected because when the material is compression molded via the one-step method, the laminate powder penetrates into the porous wet-lay material. Therefore, a more conductive network may be generated in the through-plane direction using the one-step molding method.

It is important to realize that graphite filled polymer composite plates may have different properties and performances in different directions. It is apparent from Tables 1 and 2 that the through-plane conductivities for the wet-lay based and laminate composites are significantly lower than the in-plane conductivities. This may be attributed to several possible factors. First, the graphite particles are not spherical, but rather elongated platelets that may orient in a plane perpendicular to the direction of the compaction force during molding. This plane is designated as the X–Y (in-plane) direction as shown in Fig. 3. Furthermore, when forming the wet-lay sheets, the thermoplastic and reinforcing fibers also align in the X–Y direction and may help induce the graphite particles to orient in the same direction. It is believed that for compression molded composite bipolar plates, the higher

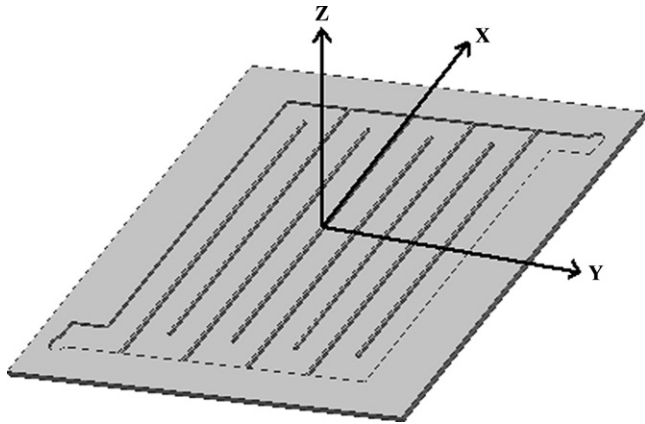


Fig. 3. Laminate bipolar plate representation showing the direction of the X–Y–Z planes.

the aspect ratio of the graphite used, the greater the ratio of in-plane to through-plane conductivity.

3.3. Mechanical properties of laminate composite materials

In addition to electrical conductivity, the bipolar plates should also have adequate mechanical properties to be used in fuel cell stacks where they would be subjected to a constant compressive load. However, for polymer composites doped with high levels of conductive particles and/or fibers, it was difficult to obtain high conductivity and sufficient mechanical properties simultaneously. As a result the mechanical properties of most composite materials used to produce bipolar plates were still lower than the target values with the exception being the PPS based wet-lay composites (see Table 1). The PVDF based laminate material exhibited lower strength than wet-lay composite material. A decrease in mechanical properties is expected with the laminate plates because the surfaces are generated from a powder form of polymer and are not reinforced with carbon fiber. However, due to the presence of wet-lay in the core, the mechanical strengths of the laminate plates still compared well to the vinyl ester and PVDF based polymer composite materials.

As stated in Section 2.3, the laminate plates were produced by either the one-step or two-step molding method. Potentially there could be differences in the mechanical strengths of plates produced by the two methods because of differences in adhesion of the outer layer to the core composite material. In Figs. 4 and 5 are shown the flexural and tensile strength of PPS based laminate flat plaque test specimens at various vol% of laminate layer and their dependency on processing method. The strengths exhibited an increase with decreasing vol% of laminate material. This was expected because a lower vol% of laminate material resulted in a higher vol% of the stronger wet-lay in the core. The PPS based laminate flat plaque test specimens generally exhibited higher strength (about 20% higher flexural strength) than that of PVDF based laminate plaques reported by Cunningham and co-workers [19]. Notably, there did not appear to be as large of a dependency on the processing method using the PPS based laminate plates as was exhibited by the PVDF based laminate plates. This may be due to the compatibility of the polymer in both the laminate and

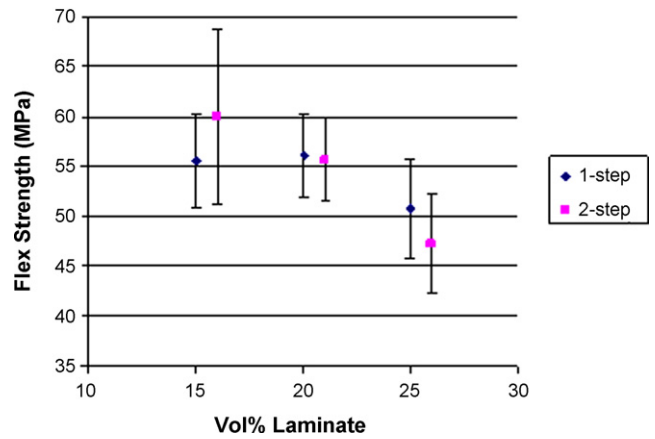


Fig. 4. Flexural strength of various volume percents of PPS based laminate layer on the surfaces of a PPS based, 70% TC-300 wet-lay core material processed by the one-step (◆) and two-step (■) methods at ambient conditions. Error bars indicate 1 S.D.

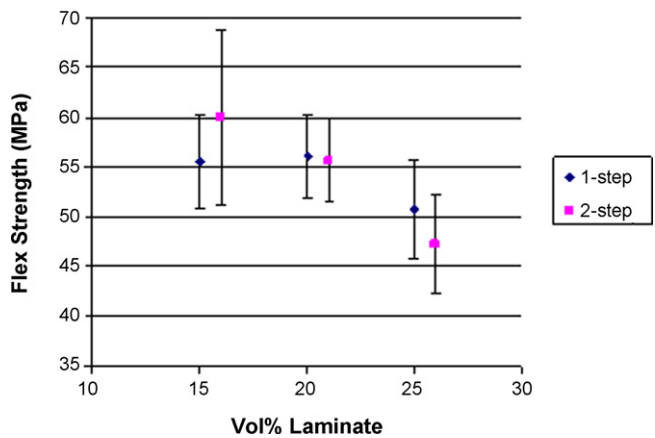


Fig. 5. Tensile strength at various volume percents of PPS based laminate layer on the surfaces of a PPS based, 70% TC-300 wet-lay core material processed by the one-step (◆) and two-step (■) methods at ambient conditions. Error bars indicate 1 S.D.

wet-lay material using PPS throughout the plate, whereas in the PVDF based laminate plates, there may not have been adequate adhesion between the laminate layer and the PPS based wet-lay core, especially in the two-step molding method. It is also believed that de-lamination could occur using a PVDF based laminate layer and a PPS based wet-lay core.

In Table 3 is shown the flexural strength and modulus of PPS based laminate flat plaques at various temperatures. Because fuel cells operate at approximately 100 °C, the properties are reported over a range of temperatures of fuel cell operation. The flexural

Table 3 Comparison of flexural strength and modulus of PPS based laminate composites at various temperatures

Material	Temperature (°C)	Flexural strength (MPa)	Flexural modulus (GPa)
15 vol% PPS laminate	23	60.0	12.0
	80	63.1	13.4
	100	60.0	7.2
	120	57.6	6.9

modulus generally decreased as temperature was increased due to the relatively low T_g of PPS. However, the flexural strength was maintained fairly well throughout the temperature testing range, measuring approximately 60 MPa, just above the DOE target value. The flexural modulus also exhibited higher values at elevated temperatures for the PPS based laminate material than that of PVDF based laminate material by approximately 30%. Therefore, in an operating fuel cell it is believed that the PPS based laminate bipolar plates will provide improved strength and stiffness over those of PVDF based laminate bipolar plates.

3.4. Half-cell resistance of laminate composite bipolar plates

Although electrical conductivity of the materials used to produce bipolar plates is important, the performance of the bipolar plate in a fuel cell is the defining characteristic. A bipolar plate may have electrical properties that drastically differ from flat plaque test specimens because of the molded gas flow channels in the surfaces. To evaluate the electrical properties of the bipolar plates, half-cell resistance tests were conducted as well as an optical analysis of the surface roughness of the bipolar plates to quantify the roughness.

In Fig. 6 are shown comparisons of half-cell resistances for 15 and 33 vol% laminate composite materials and thicknesses at an applied pressure of 220 psi. The half-cell resistances of the laminate bipolar plates with 0.5 mm deep channels were lower than those with 0.8 mm deep channels. Although the active area (surface area of bipolar plate contacting carbon paper) was not changed using shallower channels, the thickness of the bipolar plate was reduced by approximately 0.6 mm. PPS based laminate bipolar plates of 15 vol% with 0.5 mm deep channels exhibited half-cell resistance values of $0.025 \Omega \text{ cm}^2$, about 20% less than that of PVDF based laminate bipolar plates with 0.5 mm deep channels. It is believed that reducing the channel depth below 0.5 mm will allow for the total plate thickness to be further reduced, potentially lowering the half-cell resistance even more. This is demonstrated with the 33 vol% PPS based laminate bipolar plate of thickness 2.2 and 0.5 mm channel depth, which exhibited a half-cell resistance of $0.018 \Omega \text{ cm}^2$. Although 33 vol% laminate layer was not the ideal loading level for balancing electrical and mechanical properties, the dependency of

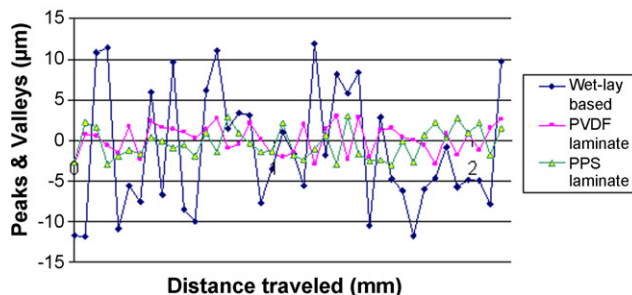


Fig. 7. Two-dimensional surface roughness profile of a wet-lay (\blacklozenge), PVDF laminate (\blacksquare), and PPS laminate (\blacktriangle) based bipolar plates.

half-cell resistance on the total plate thickness was established. Although half-cell resistance measurements were carried out under an applied pressure of 220 psi, it is acknowledged that an operating fuel cell stack may require bipolar plates to be placed under 250 psi applied pressure. Because the interfacial contact between carbon paper and bipolar plate is critical in the evaluation of half-cell resistance, and that higher pressure may improve interfacial contact, it is believed that taking measurements at 250 psi compaction pressure will show a further decrease in half-cell resistance.

3.5. Surface roughness of compression molded bipolar plates

Surface roughness is believed to significantly contribute to the half-cell resistance and performance of a bipolar plate in an operating fuel cell. Although the bipolar plate may possess good conductivity, the surface contact between the bipolar plate and the membrane electrode assembly (MEA), gas diffusion layer, etc. is important in the performance of the bipolar plate. In Fig. 7 is shown a two-dimensional surface profile of a wet-lay based bipolar plate and a PPS and PVDF based laminate bipolar plate. The PPS and PVDF based laminate bipolar plates exhibited values for peaks and valleys to reach a maximum of $\pm 3 \mu\text{m}$. The wet-lay based bipolar plate exhibited values for peaks and valleys reaching as high as $\pm 12 \mu\text{m}$. Both laminate bipolar plates exhibited significantly smoother surfaces than the wet-lay based bipolar plate. Therefore, the interfacial contact between the laminate bipolar plate and MEA should be drastically improved over that of the wet-lay based bipolar plate in an operating fuel cell. Because the surface roughness of the PVDF and PPS laminate plates were comparable, it was expected that they would provide similar interfacial contact with the MEA in an operating fuel cell.

3.6. Potential for rapid, continuous manufacturing of bipolar plates

In Fig. 8 is shown a feasible production method for the PPS based laminate material. The wet-lay material is continuously drawn through calendar rolls. It is believed that vertical consolidation will be effective for allowing the PPS based laminate powder to be metered on just before the material is consolidated into a blank. After the consolidation blank is formed it can be

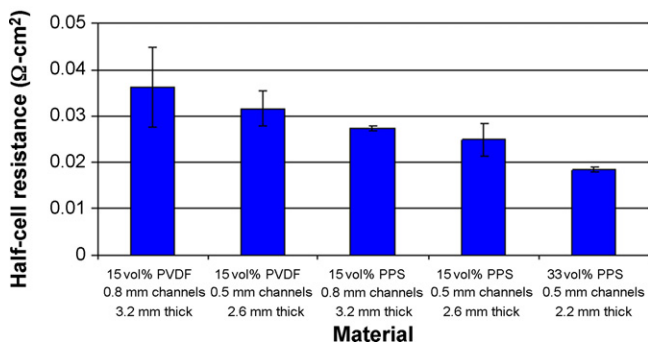


Fig. 6. Half-cell resistances of various compositions and thicknesses of laminate bipolar plates.

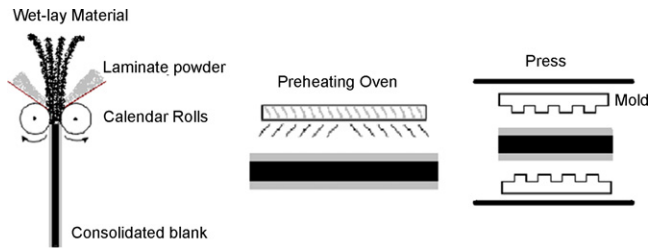


Fig. 8. Envisioned continuous process for the production of PPS based laminate bipolar plates.

cut to the desired length and width dimensions. The laminate blank can then roll underneath a heating source such as infrared radiation or induction. Once the entire blank is heated to a PPS processing temperature of 300 °C, the preheated material can then drop into a cold mold. The cold mold forms channels into the surfaces of the laminate blank while allowing the part to cool down through 230 °C, the peak crystallization temperature of PPS measured by DSC. The bipolar plate can then be removed from the mold.

4. Conclusions

A new method has been developed to produce potentially cost-effective bipolar plates with high electrical conductivity, good mechanical properties and potential rapid manufacturability. Composite sheets consisting of graphite particles, PPS fibers, carbon fibers, and microglass were first generated by means of a wet-lay process. The porous sheets were then stacked and covered with PPS/graphite particles and compression molded to form layered composite bipolar plates with gas flow channels. Compared to PVDF based laminate plates, the PPS based laminate plates exhibit higher mechanical strength, lower half-cell resistance, comparable formability, and the potential to avoid adhesion and de-lamination issues. Reducing the channel depth from 0.8 to 0.5 mm has also shown to significantly decrease the half-cell resistance. It is believed that reducing the channel depth further will allow for improvements in half-cell resistance, and require less laminate layer which may help increase the overall strength of the plate. Although the mechanical properties of the laminate plates are not as high as that of the wet-lay based plates, they are still higher than that of other polymer composite bipolar plates due to the presence of a strong wet-lay material in the core. While the one-step method is good for batch production of PPS based laminate bipolar plates, the two-step method may

be chosen for continuous mass production of the bipolar plates without compromising mechanical or electrical properties. It is believed that increasing the carbon fiber content in the wet-lay material may help provide more strength in the core. It is also believed that using nanoclays in the laminate layer may help increase mechanical properties of the plate.

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