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Percolation threshold study of a plastic–elastomeric matrix based composite material for bipolar plates in proton exchange membrane fuel cells

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Abstract

Bipolar plate materials play an important role in designing a cost-effective fuel cell. This research aims to develop a polymer matrix based composite bipolar plate material for proton exchange membrane fuel cells. The polymers investigated are a vinyl ester resin (VER) and two elastomers including polyurethane (PU) and ethylene–propylene–diene monomer (EPDM). The VER alone and the blend of VER with either PU or EPDM makes the one-phase, one-phase blend, or two-phase blend matrices. These matrices will be incorporated within the conductive fillers including Cytec DKD carbon fiber (DKD), Asbury synthetic graphite grade 4012 flake (4012), and grade TC 307 high-surface flake (307). A percolation threshold study was conducted and two-phase blend matrix can achieve lower percolation thresholds. The composite material with 65 vol.% of two-phase blend matrix (60 vol.% VER and 40 vol.% EPDM), and 35 vol.% of conductive fillers (21 vol.% DKD, 5 vol.% 4012 and 9 vol.% 307) is found to have low electrical properties with improved mechanical properties.

Keywords

Polymer blends, PEM fuel cells, conductive material, percolation threshold

Introduction

A fuel cell is an electrochemical device that converts chemical energy from fuels directly into an electrical output. The operational cell potential is less than the reversible cell potential due to several losses associated with cell operation.¹ Therefore, bipolar plates are used to connect several single cells to form fuel cell stacks to provide the desired power.

To fulfill their functions in a fuel cell stack, bipolar plates must provide the following needs. The bipolar plate provides the structural support to membrane electrode assembly. The bipolar plate also distributes reactants and oxidants to the gas diffusion layer (GDL) and removes water product from the reaction sites through the flow channels on the bipolar plate. Therefore, bipolar plate materials must be electrically and thermally conductive. The bipolar plate material must also have low gas permeability to prevent mixture between reactants and coolant.²

There are several designative types of bipolar plate material which includes carbon-based and high-purity

graphite, metal, conductive plastic, and composites.³ Each type has its advantages and disadvantages. Composites can be the most effective and time efficient to produce from a cost aspect.

Composite bipolar plates use either thermoplastics or thermosets incorporated with conductive fillers. Processing of the composites may vary based on whether thermoplastics or thermosets are used. Examples of thermoplastics used are polypropylene,^{4,5} polyethylene,⁶ polyester,⁷ polyphenylene sulfide,^{5,8} polycarbonate,^{9–11} and polyvinylidene fluoride.^{6,12} Thermosets commonly used for bipolar plates are vinyl ester (VE),^{13,14} phenolic resin,^{4,15,16} and furan resin.^{10,12} The conductive fillers being used include

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carbon fiber, carbon powder, carbon nanoparticles, expandable graphite, and other conductive alternatives.^{16–19} The blend of different conductive fillers proved to have a synergetic behavior. When the blend consists of Cytec DKD fiber and Asbury synthetic graphite grade 4012 flake in a composite material, a higher electrical conductivity results than with the fiber or flake being used alone.²⁰

The blends of different conductive fillers do not only improve the electrical conductivity, but also reduce the amount of volume percentage of conductive fillers within the composite bipolar plate. When there is a larger volume percentage of conductive fillers within the composition, the polymer connections are weakened and the composite becomes stiffer. When there are less conductive fillers, processing of composites can be less cumbersome.

The majority of polymer materials used in composite bipolar plates have high strength and high stiffness as a result of high crystalline structures. The high crystalline structures prevent easy movement of molecules relative to each other and decrease the elastic elongation.²¹ It is becoming evident that the stiffness of the bipolar plates might be hindering the performance of fuel cell. Gasket materials are used to help seal and dissipate high clamping forces when all components are put together to form a fuel cell stack. For example, if there is no protection from the incompressible gasket layer at a GDL with a thickness of 300 μm , the GDL could have a displacement of more than 75 μm , under a clamping pressure of 1 MPa.²² The ultimate goal of this research is to investigate forming a composite material that can easily incorporate a gasket into the processing of the bipolar plate.

This research aims to develop a new composite material consisting of both plastic and elastomer components. Elastomers within a plastic material will improve sealing capabilities of the fuel cell stack. This should also lower the contact resistance between the bipolar plate and the GDL. From the aspect of mechanical properties, using a thermoset resin with an elastomer will result in a material having good strength and high toughness. In this study, VE will be used as the plastic thermoset material. VEs have limited thermal stability, where hydrolysis can occur at temperatures above 100°C.²³ This maximum operating temperature is lower than the temperature of some other commonly used resins for bipolar plates, such as epoxy and phenolic. Nevertheless, VE has a price advantage, lower density, lower coefficient of thermal expansion than epoxy, comparable tensile strength to phenolic resin, and twice the elongation% of epoxy resin.²⁴ When vinyl ester resin (VER) is modified with an elastomer, the mechanical properties improve.²⁵ When selecting particular elastomers, it is important to

make sure that the elastomers are thermally stable. Also, they must have similar cure processes to not interfere with the VE. There have been studies that have suggested that immiscible polymer blends mixed with carbon black is a promising method in reducing the percolation threshold of electrical resistivity.²⁶ Plastic with elastomer blends are yet to be investigated though with the incorporation of conductive fillers in a bipolar plate. In this study, two elastomers were investigated and incorporated within the VE. One of the elastomers investigated creates a one-phase (miscible) blend polymer system, whereas the other elastomer produces a two-phase (immiscible) blend polymer system.

When making composite bipolar plates, conductive fillers are added into matrix materials at concentration levels that are well above the point that the composite plate shows electrical conductivity.²⁷ The critical point where the materials begin to achieve electrical conductivities is known as the percolation threshold. The point of percolation threshold distinguishes between a material that is an insulator and a conductor. When infinite amounts of conductive agglomerates are formed, the composite becomes conductive.²⁸ Although the composite bipolar plates need to be well above the percolation threshold, the percolation threshold can provide insight into how different polymer phase blends will interact with the conductive fillers to achieve lower electrical resistivity at an earlier percolation threshold value.

Therefore, in this particular investigation, low volume percentages of conductive fillers are added within the matrix blends to obtain more insight into the interaction between the conductive fillers and the matrix blends. The volume percentages of conductive filler range from 2 to 15 vol.%. The conductive filler used specifically for this investigation is a high surface area synthetic graphite from Asbury Carbons – grade TC 307. The article will first discuss manufacturing of the composite samples. This is followed by discussion of results and conclusions.

Experimental methodology

There were three types of matrices investigated: a polyvinyl ester producing a single-phase matrix, a compatible single-phase blend of polyvinyl ester (VER) (Reichhold Hydrex 33350-00) and a polyurethane (PU) elastomer (Eager Polymers EP1150), and a two-phase system of polyvinyl ester and an ethylene-propylene-diene monomer (EPDM) (Trilene[®] 65 Liquid Polymer). Table 1 presents the material properties of the matrix materials used in this research. Both VER and PU can be cured at room temperature, whereas EPDM is to be cured with peroxide that

requires temperatures to be elevated. The particular urethane being used is a two-part component system that does not require any additional cure agents. The two cure agents are dicumyl peroxide on a carrier of calcium carbonate (Di-Cup® 40°C) and methyl ethyl ketone peroxide (MEKP). The cure agents were found to have little influence on the electrical or mechanical properties of the samples. The only sufficient difference between the two cure agents is the amount of processing time required. DiCup only requires approximately 30 min for the cure process to be completed, whereas MEKP requires at least 24 h and additional time for post-cure. Therefore, only DiCup will be used as the cure agent in this study.

There are three conductive fillers being used within the scope of this study, Cytec DKD fiber,²⁹ Asbury synthetic flake like powder, grade number 4012,³⁰ and Asbury synthetic graphite with a high surface area, grade number TC 307. Properties of these conductive fillers are summarized in Table 2.

All the matrix materials selected have a similar processing procedure, since they are all liquid polymers. Processing consists of mechanical mixing of the matrix and incorporating the cure agent. After the matrix material is mixed well with the other polymer and cure agent, the conductive fillers are then added to the mixture. After all the components are thoroughly mixed, the mixture is placed in a compression mold for 30 min at 170°C under a compression of 35 MPa.

Some additional preparations are taken with the use of EPDM. The EPDM has a high molecular weight, which drives the viscosity up and makes it difficult to process the blends. The manufacturing process, however, can be modified since EPDM has good structural design adaptability.³¹ To make processing easier, the EPDM was heated up in a flat-bottom mantle heater with a controlled temperature of 90°C. When the urethane was mixed with VER, preprocessing does not require heating up the mixture before placing the composition in the compression mold. If the two components are mixed together, they will eventually cure at room temperature. The introduction of DiCup and VER will only speed up the curing time. DiCup does not interfere with curing process of the urethane.

Once samples are made, both electrical and mechanical properties were measured. An MTS Insight 50 was used for testing of both tensile and flexural properties. The procedure for tensile testing followed ASTM D 638 closely. The test procedure for flexural strength followed ASTM D 790 closely. Electrical resistivity was measured with a four-point probe apparatus by Signatone. There are four probes, with the inner two measuring the voltage and outer two the current. The resistance is measured with a Keithley 195A digital multimeter. The electrical resistivity was found using correction factors to account for the probe spacing and the sample dimensions which was developed by Smits³² in 1957. Equation (1) shows how to calculate the electrical resistivity using the resistance measured and the overall dimensions of the samples.

$$\frac{\rho}{t} = R_c = CF_1 \times CF_2 \times R_M \tag{1}$$

where ρ is the electrical resistivity, t the sample thickness, CF1 and CF2 the two correction factors which account for the width and length (D) and thickness (t) of the sample relative to probe spacing (s), R_c the corrected resistance, and R_M the measured resistance from the Keithley.

Table 1. Matrix material properties

Property	VE	EPDM	Urethane
Density (g/cm ³)	1.08	0.86	1.04
Hardness	44 BH	65 A	50 A
Tensile strength (MPa)	83	–	2.2
Flexural strength (MPa)	145	–	–

Table 2. Conductive filler selected properties

Property	Cytec graphite fibers DKD	Asbury graphite	
		Synthetic flake 4012	Synthetic high surface area graphite TC 307
Density (g/cm ³)	2.2	2.23	2.16
Electrical resistivity (Ω-cm)	<3 × 10 ⁻⁴	0.03	0.24
Thermal conductivity (W/m-K)	400–700	–	–
Diameter (μm)	10	50–100	1–2 (width)
Length (μm)	200	40–300	10–100 (thickness)
Surface area (m ² /g)	0.4	1.5	352

Results and discussion

The first portion of the testing was performed in this research to determine how electrical resistivity is affected by different ratios of elastomers to thermosets. Table 3 presents a summary of all the matrix ratios that were tested for the two-phase blend (EPDM-VER), as well as one-phase blend (urethane-VER) with 35 vol.% conductive fillers. Figure 1 compares the electrical resistivity of composite samples made from one- and two-phase blends. Each blend contains 65 vol.% matrix and 35 vol.% conductive filler (DKD and 4012). The ratio of VER increases from 10% to 100% in each matrix blend. As the

concentration of elastomers in both one- and two-phase blends are increased, the electrical resistivity is increased. The elastomers are more electrically and thermally insulating than the VER. Therefore, the electrical resistivity of a composite material can be hindered depending on how the conductive fillers are situated within the composition.

The mechanical properties also demonstrate some correlation to electrical resistivity. Figure 2 shows the experimental results under the tensile loadings. As shown in Figure 2, the differences in mechanical energy absorption or strain energy density are evident by integrating the stress-strain curves of different matrices. The strain energy density is represented by patterned lines in Figure 2. The composite material containing two-phase blend (40 vol.% EPDM to 60 vol.% VER) within the 35 vol.% conductive fillers has the most significant amount of mechanical energy. The overall strain energy density for the two-phase blend containing 40 vol.% EPDM to 60 vol.% VER is 0.24 MN m/m^3 as compared to 0.09 MN m/m^3 for a one-phase blend containing 40 vol.% PU to 60 vol.% VER PU/VER and 0.03 MNm/m^3 for VER alone. Figure 3 shows the experimental results under the flexural loadings. Improved elongation of samples was observed when either PU or EPDM was present. Strain of samples was about 1.5 times or more greater when elastomer was present than that when only VE was used.

Table 3. Matrix volume ratios of blends within 35 vol.% of conductive fillers

Matrix volume ratio	
VE:EPDM	VE:Urethane
100:0	100:0
70:30	90:10
60:40	70:30
40:60	60:40
	50:50
	40:60
	10:90

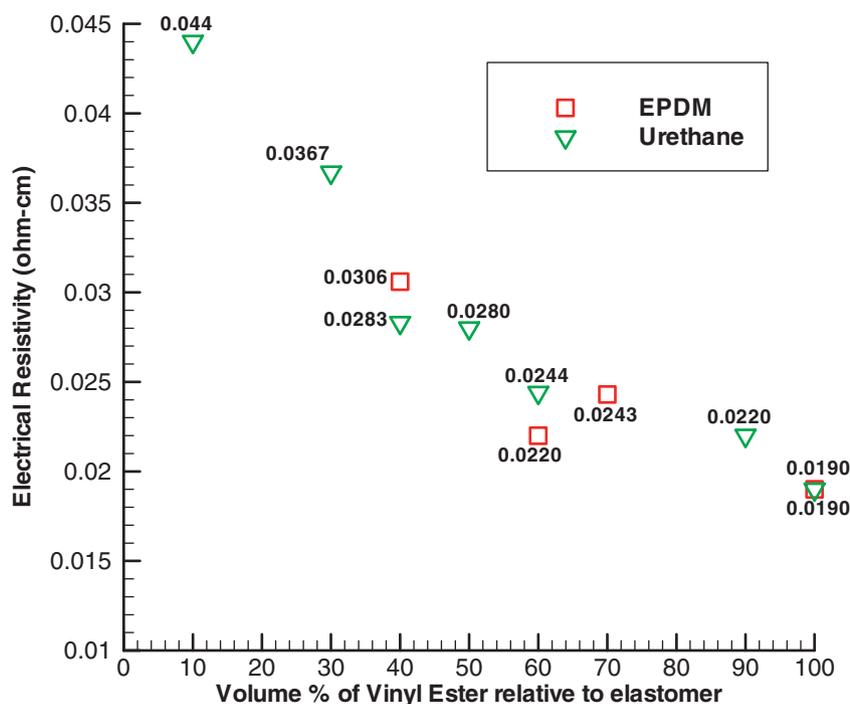


Figure 1. Electrical resistivity based on VE-to-elastomer ratio with a 35 vol.% of conductive fillers (DKD-4012).

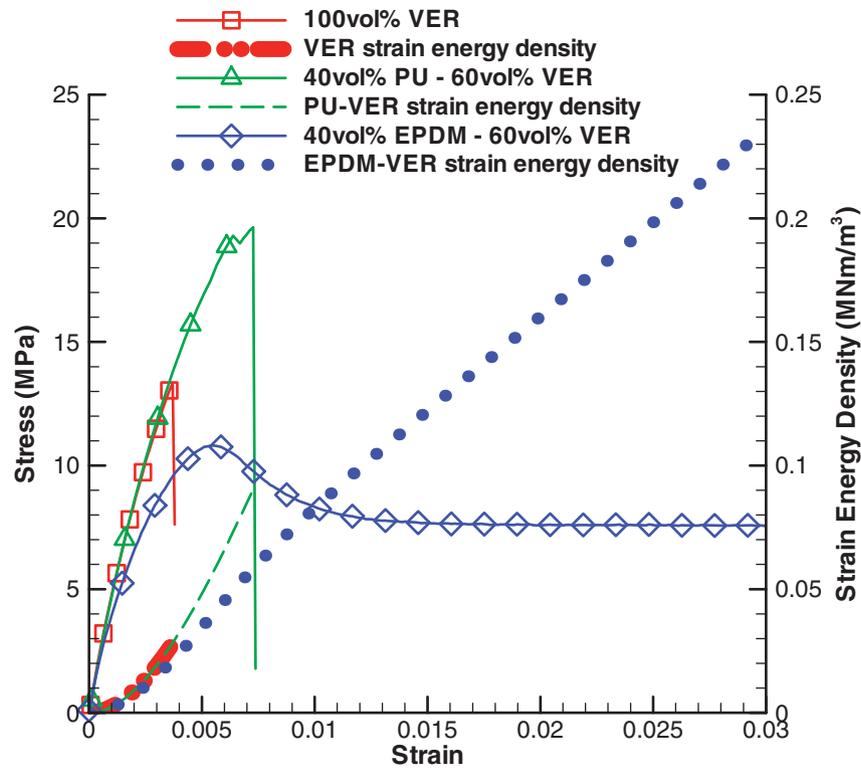


Figure 2. Stress–strain relationship comparing EPDM, PU, and VER matrices within 35 vol.% conductive fillers (DKD–4012) under a tensile loading.

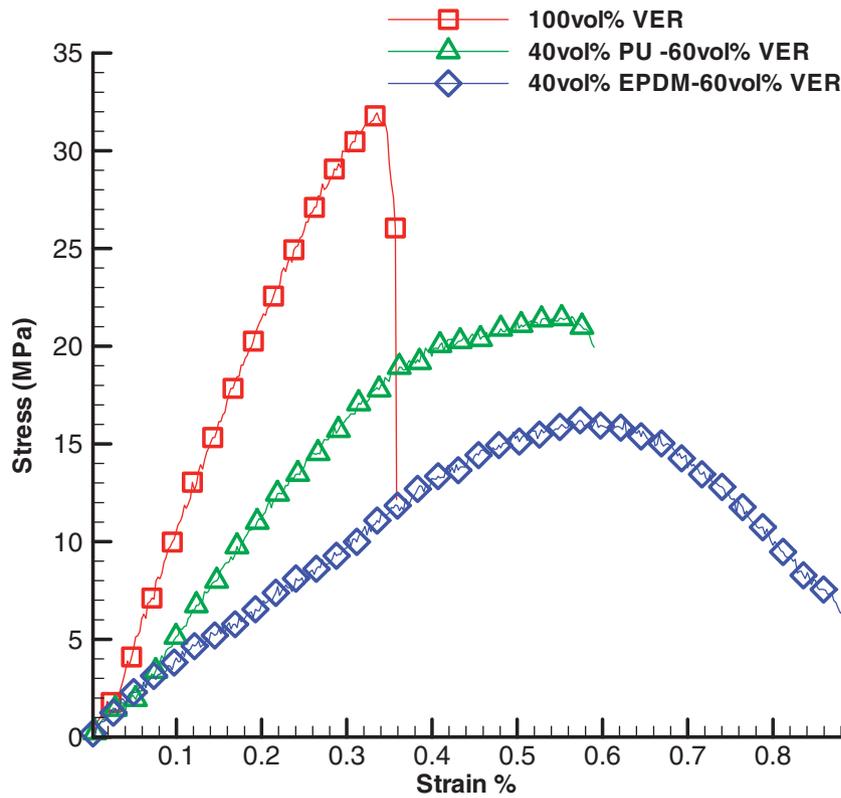


Figure 3. Stress–strain relationship comparing EPDM, PU, and VER matrices within 35 vol.% conductive fillers (DKD–4012) under a flexural loading.

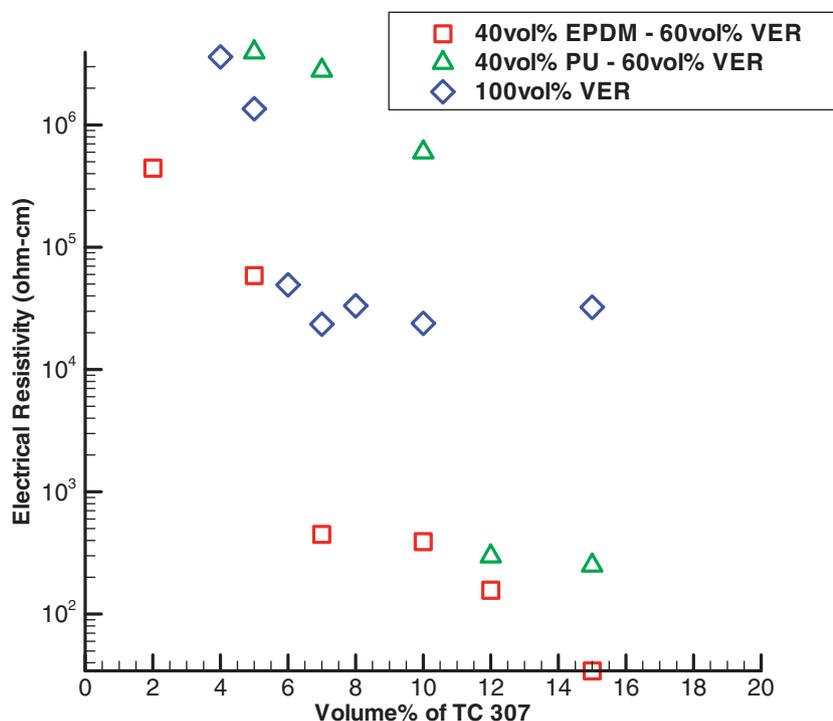


Figure 4. Electrical resistivity of percolation threshold conductive fillers added to different blend matrices of elastomer and VE.

To further understand how the conductive fillers are behaving within the compositions, very low percentage of conductive filler is used. The electrical resistivity will show more dramatic changes due to the pathways of conductive filler starting to form. This test will also help explain the mechanical behavior of the one phase and one and two-phase blends.

The experiment adopted a nano-scale ultra high surface area synthetic graphite made by Asbury Carbons. The grade of the material is referred as TC 307. Since the composite with a ratio of 40 vol.% elastomer to 60 vol.% of VER showed more elastic behavior and relatively low electrical resistivity for both the one- and two-phase blends, these ratios were used to perform the percolation threshold study on TC 307. We added TC 307 to the different matrix blends at volume percentages ranging from 2% to 15%. Figure 4 shows the electrical resistivity of the composite material we have made. The matrix that consists of the two-phase blend achieves levels of percolation at a 7 vol.% of TC 307, which is earlier than the one-phase blend matrix at a 12 vol.% of TC 307. VER alone had an initial percolation around 7 vol.% of TC 307 same as the two-phase blend; however, the electrical resistivity was approximately two orders of magnitude higher. A focused ion beam was used to clean up the rough topology in order to observe the nano-fillers. The ions were used in a high vacuum chamber to hit a precise location on the part of the sample that was being viewed under the scanning electron microscope (SEM).

Since VER (one phase) and EPDM-VER (two-phase blend) achieved signs of percolation at 7 vol.% of TC 307, they were both observed under an SEM, shown in Figure 5(a) and (b), respectively. The images are scaled so that the nano-fillers in theory should be about the same size in each image. Nano-fillers in the localized SEM image of Figure 5(a) are in more of an agglomerate configuration than they are in Figure 5(b). This explains the electrical resistivity being approximately two magnitudes higher than that of the two-phase blend system. The nano-fillers are able to distribute more within the two-phase blend. This also supports that the hypothesis that the fillers are situating themselves at the interphase between the VER and EPDM, allowing a continuous path at a lower concentration.

Figure 6 compares the SEM images of the EPDM-VER (two-phase blend) with PU-VER (one-phase blend). The composite material with one-phase blend matrix contains more agglomerates, as shown in Figure 6(b). There are also more fillers within the one-phase blend since the percolation threshold was not observed till a 12 vol.% TC 307 was added to the composition. Even with the addition of 15 vol.% TC 307 within the one-phase blend, the electrical resistivity was not lowered significantly. This explains that although there may be localized spots of low electrical resistivity, the agglomerates leads to poor distribution.

Based on the findings from the percolation threshold results, if the nano-fillers are going to be added to the

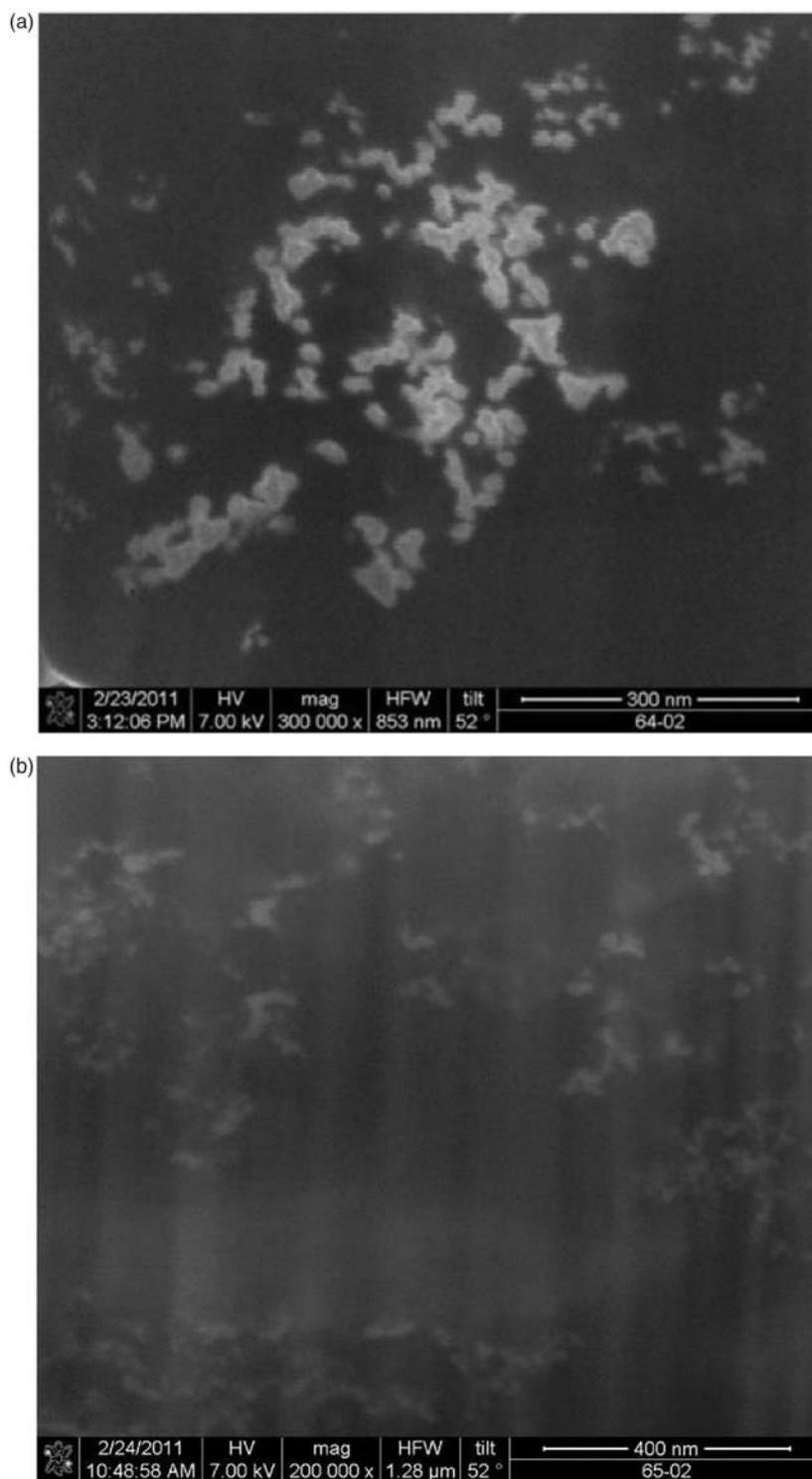


Figure 5. Cross-sectional view of (a) VER with 7 vol.% TC 307 and (b) EPDM 40 vol.% relative to 60 vol.% VER with 7 vol.% TC 307.

system with the 35 vol.% conductive filler, there may be a decrease in the electrical resistivity. In order to determine whether 7 vol.% of TC 307 was the ideal amount of nano-fillers for the composite containing a total of 35 vol.% of conductive filler, different ratios close to the percolation threshold were also evaluated. The different

ratios of TC 307 were between 6 and 10 vol.% relative to the DKD-4012 ratio that was formed due to the synergetic study. The ratio of DKD-4012 was kept constant from our previous studies²⁰ with 80 vol.% DKD relative to 20 vol.% of 4012. The results of this combined study are shown in Figure 7. It can be observed

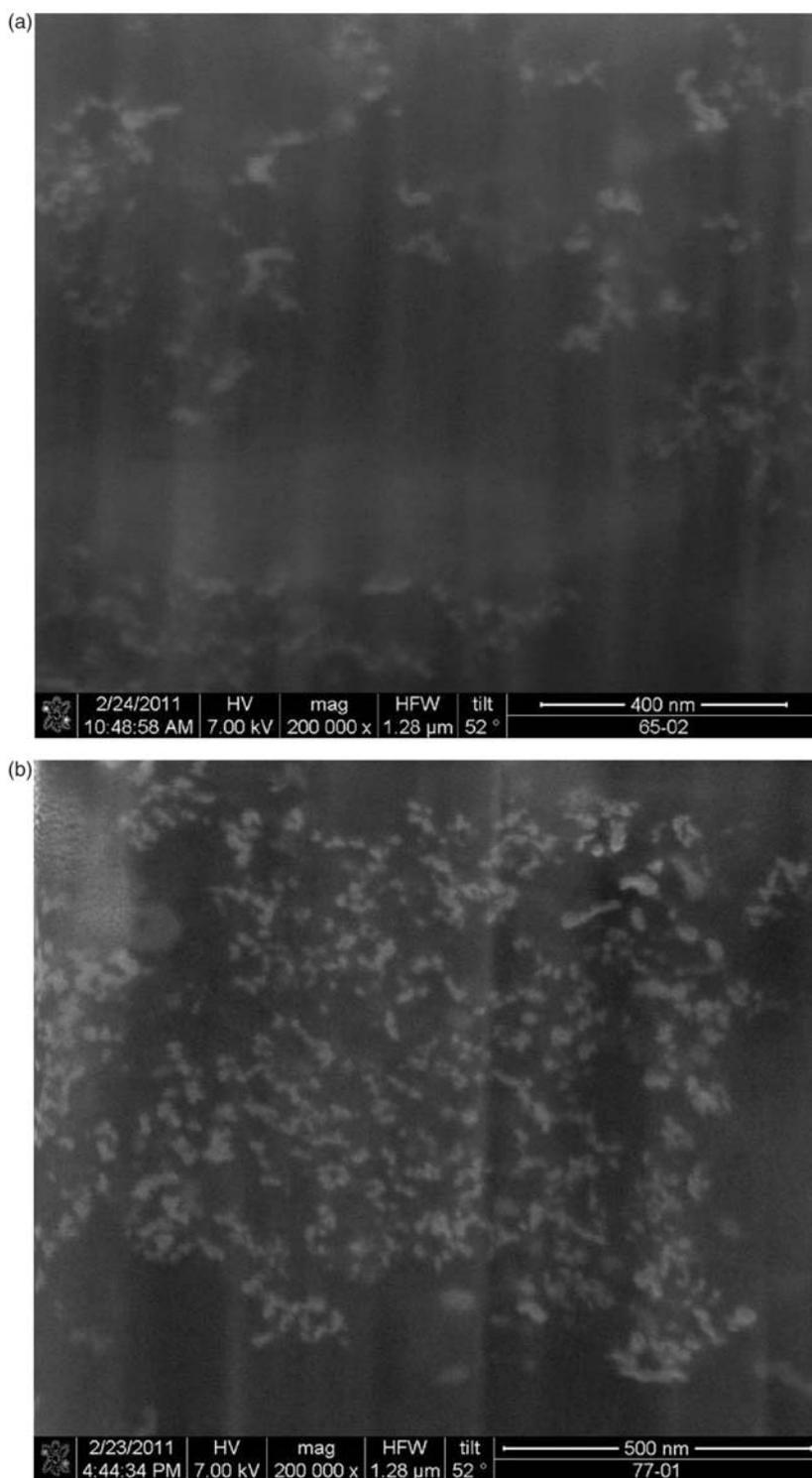


Figure 6. Cross-sectional view of (a) EPDM 40 vol.% relative to 60 vol.% VER with 7 vol.% TC 307 and (b) PU 40 vol.% relative to 60 vol.% VER with 12 vol.% TC 307.

that when TC 307 was added to the system, the electrical resistivity decreases even more from when no TC 307 is present within the composite. The sample had the lowest electrical resistivity of $0.0099 \Omega\text{-cm}$ when

containing 9 vol.% of TC 307 and 26 vol.% of DKD and 4012 at a ratio of 4 to 1. This low resistivity meets the requirements set by the design of experiment (DOE) for the electrical resistivity for composite bipolar plates.

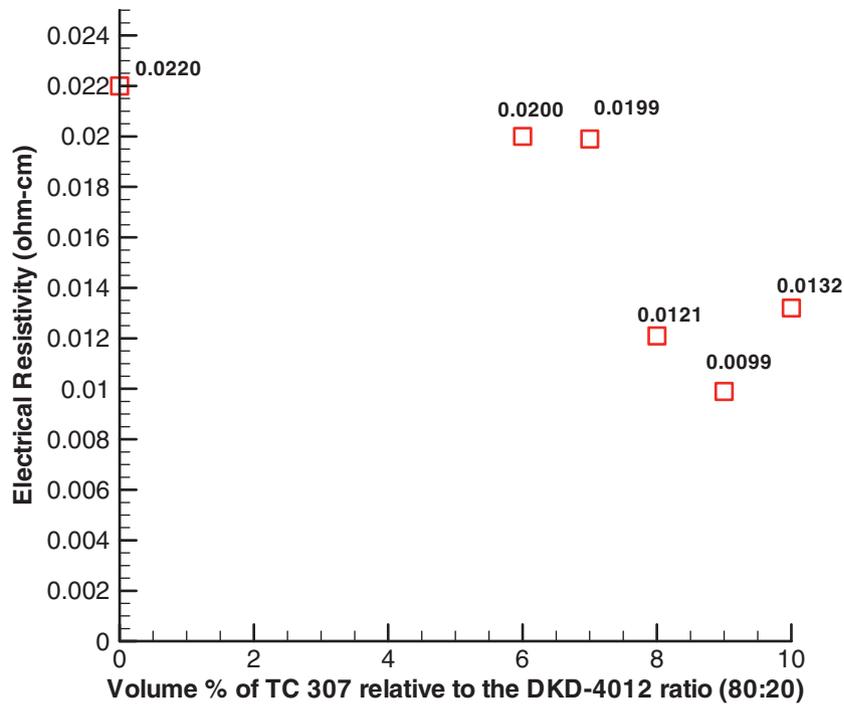


Figure 7. Electrical resistivity with incorporating TC 307 within 35 vol.% conductive fillers and the two-phase blend of VE and EPDM.

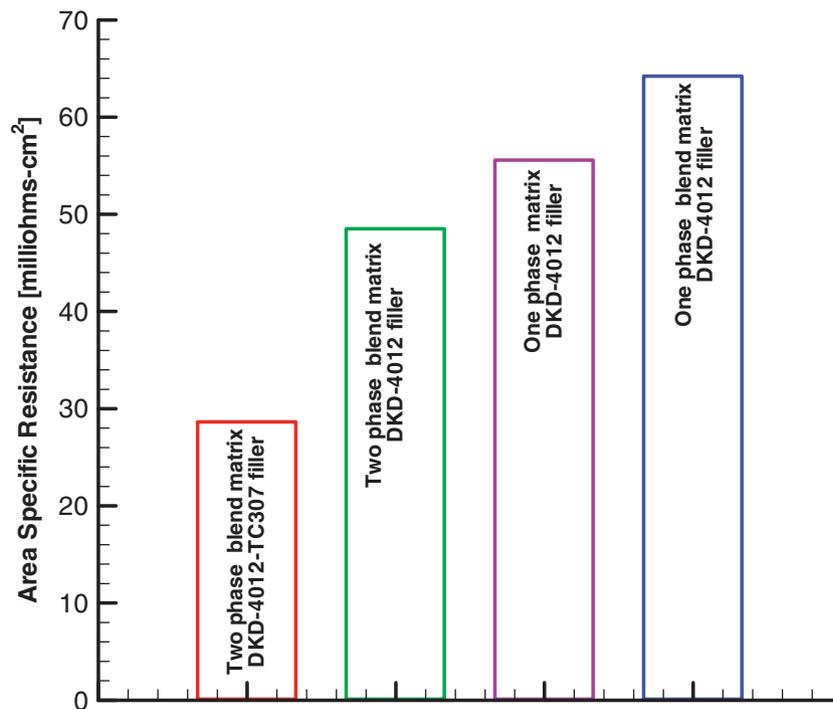


Figure 8. Comparison of area-specific resistance of different blended matrices with conductive fillers.

Through-plane electrical resistance of the composite bipolar plate was determined under a 1.5 MPa compression pressure. Figure 8 shows the testing results. Among the samples of one phase and one- and

two-phase blends, the sample containing the 9 vol.% TC 307 within the 35 vol.% of conductive fillers in the two-phase blend of EPDM and VER, had the lowest electrical resistance. Therefore, the through-plane

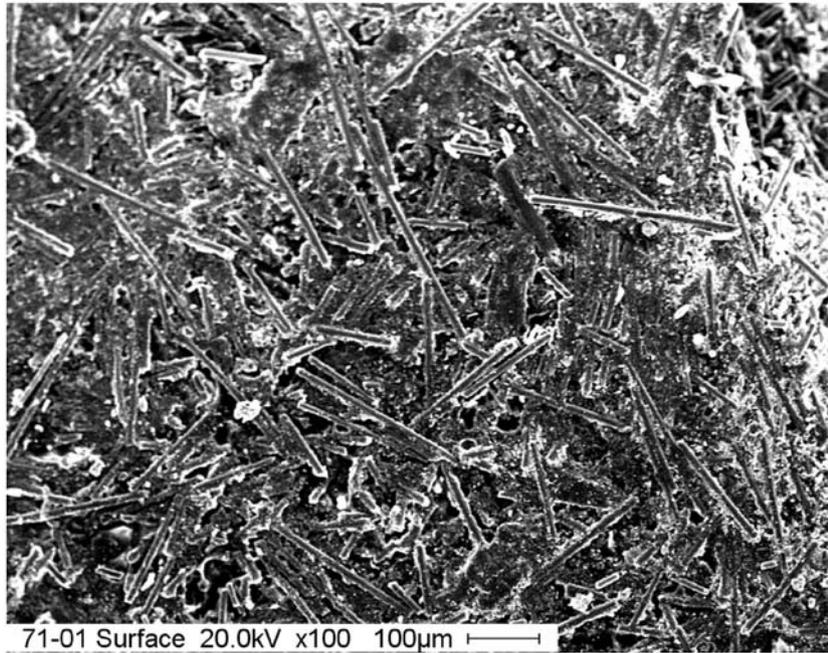


Figure 9. Surface view observed under the SEM of 40 vol.% EPDM relative to 60 vol.% VER with 9 vol.% TC 307 within the 35 vol.% of conductive fillers (DKD-4012).

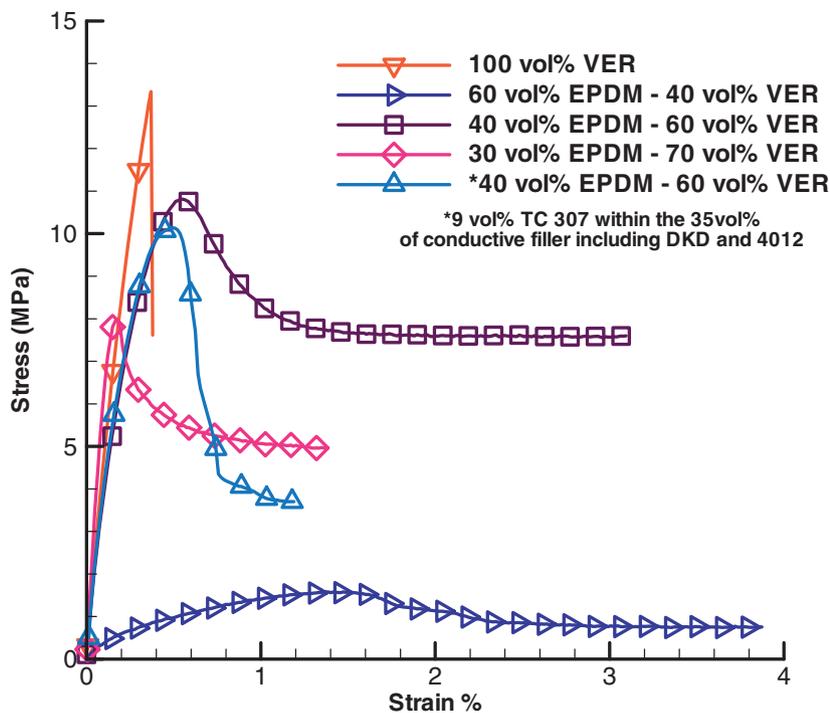


Figure 10. Stress–strain relationship of different ratios of EPDM to VER and the effects of the TC 307 (9 vol.% of the total 35 vol.% conductive fillers) present under a tensile loading.

electrical resistance follows the same trend as the in-plane electrical resistivity measurements. Figure 9 shows the surface view of the optimal composition created with the 40 vol.% EPDM relative to 60 vol.%

VER matrix and the 9 vol.% TC 307 within the 35 vol.% of conductive fillers.

Conductive fillers in a composite material expect to influence the mechanical properties of the composite

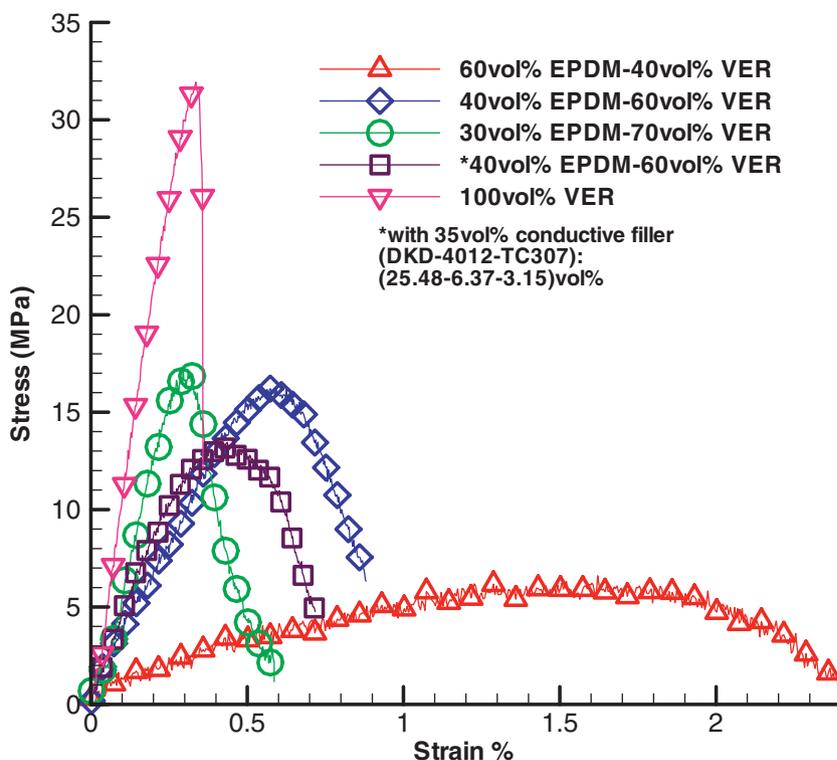


Figure 11. Stress–strain relationship of different ratios of EPDM to VER and the effects of the TC 307 (9 vol.% of the total 35 vol.% conductive fillers) present under a flexural loading.

Table 4. Comparison of compression modulus within different two-phase blend matrices and conductive fillers

Relative vol.% of matrix blend		Conductive filler vol.% of a total 35 vol.%			Compression modulus (MPa)
EPDM (vol.%)	VER (vol.%)	DKD (vol.%)	4012 (vol.%)	TC (vol.%)	
60	40	28	7	–	15
40	60	28	7	–	35
40	60	26	6.5	2.5	26
40	60	25.48	6.37	3.15	23
0	100	28	7	–	400

sample. To further understand this effect, both tensile and flexural strengths were investigated. All the tested samples contain 35 vol.% conductive fillers and 65 vol.% matrix. The matrix material is a two-phase blend of EPDM and VER. Figures 10 and 11 show the tensile and the flexural loadings, respectively, of the two-phase blend matrices with the 35 vol.% conductive fillers. For the tensile loading, it can be observed in Figure 10 that the addition of the nano-fillers with the other conductive fillers allowed the two-phase blend matrix system to withstand similar stresses without compromising the elastic quality of the composition. For the flexural loading, it can be observed that the addition of nano-fillers makes the material not able to withstand quite as much deflection; however, it is

well within the requirements of flexibility for the composite bipolar plates set by the DOE. Table 4 presents the comparison of the different compression moduli for the compositions. The compression modulus showed good elastic behaviors when EPDM was present within the composition. When only VER was present within the composition, the compression modulus was high, as expected for brittle-type polymer.

Conclusion

Both single (VE and urethane) and two-phase (VE and EPDM) blends of matrix material have been used to make a composite material, which can be used for bipolar plate in a proton exchange membrane fuel cell.

It has been observed that depending on blends of elastomer and thermosets, mechanical properties as well as electrical resistivity properties can be dramatically affected. When only VE was present within conductive fillers, the material was observed to be very stiff. When elastomers were added, the toughening of the VE was observed, however, until the elastomers were forming the continuous phase of the matrix material, and the compositions appeared to still have a plastic behavior. Blends with EPDM consistently showed signs of making the compositions more durable and also appear to allow conductive fillers to network between the matrix materials at lower percolation values. Allowing conductive networks to form thus helps reduce the electrical resistivity.

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Conflict of interests

None declared.

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