

Electrical properties of carbon-based polypropylene composites for bipolar plates in polymer electrolyte membrane fuel cell (PEMFC)

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Abstract

An investigation is made of the electrical properties of polypropylene/graphite (PP/G) composites as prospective replacements for the traditional graphite bipolar plate in proton-exchange membrane fuel cells. The composites have relatively low electrical conductivities, i.e., up to 28 S cm^{-1} at 90 wt.% G. Combination of G with carbon black (CB) is an effective way to develop higher conductivity composites. The conductivity reaches 35 S cm^{-1} by combination of 25 wt.% CB and 55 wt.% G to 20 wt.% PP. This is five times the value at 80 wt.% G and 20 wt.% PP (7 S cm^{-1}). Two methods are mainly adopted for the preparation of composites, namely, melt compounding and solution blending. Solution blending of PP with conductive fillers followed by moulding of the dried powder leads to higher conductivities compared with those of melt-compounded composites. The combination of conjugated conducting polymers such as polyaniline (PANI) with the PP, G, and CB is also investigated. It is found that composites containing PANi have lower conductivities than those of the neat composites. This decrease in conductivity is attributed to the poor thermal stability of PANi.

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1. Introduction

Considerable attention has been given to the synthesis of conductive polymer composites (CPCs) and to studies of their properties to as replacements for traditional graphite bipolar plates in proton-exchange membrane fuel cells (PEMFCs) [1–6]. These materials have low density and good corrosion resistance in combination with good electrical conductivity that can be varied over a wide range. Common conductive fillers such as carbon fibre (CF), graphite (G), and carbon black (CB) have been introduced to polymeric materials to change their electrical properties to meet the optimum design requirements for bipolar plates.

Recent literature has showed some interesting progress in polymer/G composite bipolar plates [7–13] of which some are commercially available. The main processes to produce these plates are compression moulding and injection moulding. Most compression moulding methods start with a powder compound. This powder is fed into a heated mould in which the compound

will flow and fill the mould cavity. If a thermoplastic binder is used, the mould has to be cooled to a temperature below the melting temperature of the binder before the plate can be removed. The electrical conductivities of some polymer/G composites are still below required that for bipolar plates. Polypropylene/G composites (PP/G) are among the polymer/G composites that have been used to produce bipolar plates [14–16]. Several methods of processing have been developed for composite bipolar plates. Polypropylene is one of the most widely used polyolefin polymers that can form dry mixtures suitable for compression moulding when combined with G powder.

Because PP does not include any polar groups in its backbone, it is thought that homogenous dispersion of the graphite layers in PP is not realized and thus a relatively low electrical conductivity is obtained. Moreover, hardly any interaction occurs between PP and chemically inert G layers [17]. Combination of G with other conductive fillers such as CB has been shown to be an effective way to develop higher conductivity PP/G composites [18,19]. Carbon black is an additive characterized by a smaller particle size (10–50 nm) than graphite. Combination of graphite powder with a minor weight fraction of CB introduces a synergistic effect, which results in a significantly higher conductivity than

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with single G filler at the same filling degree. The conductive network built up by the binary filler system is assumed to consist of highly dispersed CB particles bridged by G particles. Additionally, CB presents a greater tendency to build up highly structured aggregates and agglomerates that increase the probability of contacts. Aggregates have a tendency to form network structures at low concentration of the filler depending on the rheology of the polymers during the mixing process and in post-processing step, the wettability of particles by the polymer, solidification rates after mixing, the degree of crystallinity of polymeric matrix, and polymer–carbon black interactions [20–23].

Less attention has been given in the literature to the use of conjugated conducting polymers such as polyaniline (PANI) and polypyrrole (PPy) as conductive fillers in composite bipolar plates. Among the conjugated conducting polymers, PANi is known as having probably the best combination of stability, conductivity and low cost [24–29]. Methods to produce PANi containing composites [30–32] are classified into two groups: (i) synthetic methods-based on aniline polymerisation in the presence of, or inside, a matrix polymer and (ii) blending methods to mix a previously prepared PANi with a matrix polymer. The use of PANi has been reported only for corrosion resistance and conductive coatings for metallic bipolar plates [33]. PANi is a hydrophilic conductive additive that has high surface free energy, while the base polymer PP is hydrophobic and has relatively low surface free energy. Thus, polar PANi is likely to migrate towards the interface between PP and G so that it is expected to fill the insulating gaps between the G particles and hence improve the electron transport from G to PP. To improve the miscibility of PANi and PP, dodecyl benzenesulfonic acid (DBSA) has been used as a dopant and plasticizer for PANi and it may also assist the compatibility of PANi and PP.

Two methods are mainly adopted in this work, namely, melt compounding and solution blending, for the preparation of composites to explore further the mechanism of conduction in composites regardless of the optimum formulation or the optimum processing conditions required to produce bipolar plates for PEMFCs. Both methods have advantages and disadvantages: the use of organic solvents in solution blending is not desirable because of health and environmental protection concerns while it is rather difficult to obtain a homogenous material by a dry-mixing process. The morphologies of PP/G composites as well as the thermal stabilities of the PANi composites are also investigated.

2. Experimental

2.1. Materials

Synthetic graphite powder used in this study had a density of 1.8 g cm^{-3} an electrical resistivity of $1295 \times 10^{-6} \Omega \text{ cm}$, and an average particle size of $10 \mu\text{m}$. It was supplied by GME Carbon Sdn. Bhd., Malaysia. Carbon black N330 (HAF) was also obtained from Cabot Sdn. Bhd., Malaysia, with the following specifications: 30 nm particle size, $1.7\text{--}1.9 \text{ g cm}^{-3}$ density, $0.341 \Omega \text{ cm}$ resistivity, and $254 \text{ m}^2 \text{ g}^{-1}$ surface area. Polypropylene (PP) grade Titan (600), 910 kg m^{-3} and $10 \text{ g}/10 \text{ min}$, was

supplied by Polypropylene Malaysia Sdn. Bhd. For the synthesis of PANi, the following materials were used. Aniline (Fisher, 99.8%) was vacuum distilled and stored at 3°C before use. Ammonium peroxydisulfate, APS (Fisher, 98%), DBSA (mixture of C10–C13 isomers, Fisher), hydrochloric acid (Merck, 37%), and methanol were used as received. *p*-Xylene pure (Fisher, 99%) was used in the solution blending method.

2.2. Synthesis of PANi

A processible PANi, doped and complexed with DBSA, was synthesised by one-step oxidative polymerization of aniline as follows [34,35]: Aniline (19 g) was added with stirring to a flask (located in an ice bath) containing 200 ml of 1 M HCl and DBSA in a selected mass ratio of DBSA:aniline = 0.25. The oxidation occurred by dropwise addition of APS (47.5 g), in 300 ml of 1 M HCl to the aniline solution. This was conducted with stirring for 1 h at a temperature controlled around 5°C , with further stirring overnight at room temperature. A dark-green PANi suspension was obtained and then filtered and rinsed with methanol and distilled water (20:80, v/v) until the washing liquid was completely colourless. The wet product composite was dried under vacuum at 80°C for 6 h. This pathway was selected because it was expected that the aniline-DBSA salt moieties of the polymer would enhance its solubility, and hence the processibility, while the aniline hydrochloride acid salt moieties would provide a good conductive structure for the resulting polymer. The conductive salt exhibited an electrical conductivity of around 5 S cm^{-1} as a pressed pellet.

2.3. Preparation of composites

2.3.1. Melt compounding

The components of the composites, PP and conductive filler/fillers (i.e., G, CB, and PANi), were mixed manually in a small bucket before being melt compounded in an internal mixer with a Haake torque rheometer at a temperature of 175°C . The rotational speed and the mixing time were set at 35 rpm and 10 min, respectively.

2.3.2. Solution blending

The polymer (PP) was dissolved in xylene at 140°C and the conductive filler/fillers were intimately mixed with the solvent–polymer solution. After thoroughly mixing the filler/fillers with the solvent–polymer solution for 30 min, the solvent–polymer solution containing the dispersed filler/fillers was cooled to room temperature and left until the solvent was evaporated. The resulting hardened, solid polymer which, contained filler/fillers that were substantially uniformly dispersed, was washed with methanol and vacuum dried.

2.4. Compression moulding

The composites, which were obtained by melt compounding or solution blending, were pulverized to form a powder. The powder was put in a mould, preheated in a hot pressing machine for 10 min, and then hot pressed into discs of 25 mm diameter

and 2 mm thickness at a temperature of 200 °C and a pressure of 75 kg cm⁻² for 3 min for the purpose of electrical conductivity measurements.

2.5. Characterization

2.5.1. DC conductivity measurements

The conductivities of the discs in the plane direction were determined by means of a Jandel Multi Height Four-Point Probe combined with a RM3 Test Unit, which had a constant-current source and digital voltmeter designed especially for the four-point measurements. The technique measured sheet resistance in the range from 1 mΩ/sq up to 5 × 10⁸ Ω/sq and a volume resistivity range from 10⁻³ to 10⁶ Ω cm. The system accuracy was within 0.3%.

2.5.2. Thermal analysis

A Mettler, Toledo thermogravimetric analyzer (Model TGA 851e) was used to determine the thermal stabilities of the PANi composites. The specimens in this test were scanned from 25 to 600 °C at a heating rate of 20 °C/min in the presence of nitrogen.

2.5.3. Morphological observations

The polished surface morphologies of the composites were investigated with a variable-pressure scanning electron microscope (VPSEM, Model LEO 1450VP) at an accelerating voltage of 20 kV. The samples were gold-sputtered prior to SEM examination. Backscattered electrons were utilized to provide atomic number contrast between the composite components.

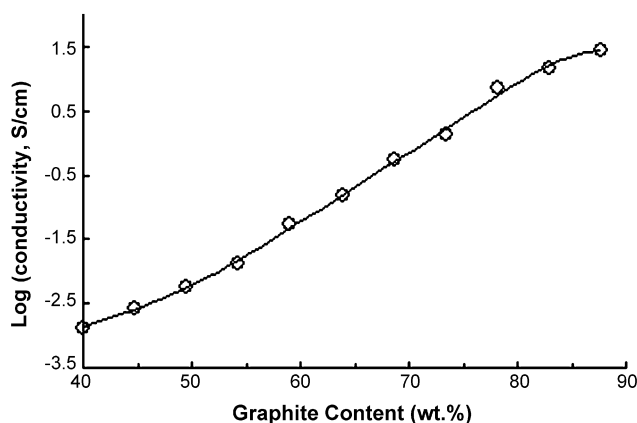


Fig. 1. Log conductivity as function of G content for PP/G composites.

3. Results and discussion

3.1. Electrical conductivity

3.1.1. PP/G composites

The absolute values of the electrical conductivities of all the composites are summarized in Table 1. The dependence of the electrical conductivity of the PP/G composites on the filler content is shown in Fig. 1. The reported values represented the average of five readings. The results show the expected trend of an increase in conductivity with an increase in filler loading. The conductivity is raised from 10⁻¹⁶ S cm⁻¹ (for neat PP) to ~10⁻³ S cm⁻¹ for PP with 40 wt.% G. As the filler

Table 1
Absolute values of conductivities of PP/G, PP/G/CB, PP/G/PANi and PP/G/CB/PANi composites

G (wt.%)	CB (wt.%)	PANi (wt.%)	σ (S/cm), Melt compounding	σ (S/cm), Solution blending
40	0	0	1.33×10^{-3}	6.63×10^{-2}
50	0	0	5.84×10^{-3}	1.88×10^{-1}
60	0	0	5.37×10^{-2}	8.15×10^{-1}
70	0	0	5.37×10^{-1}	4.89×10^0
80	0	0	7.04×10^0	2.32×10^1
35	5	0	5.70×10^{-3}	3.20×10^{-1}
45	5	0	3.70×10^{-2}	1.02×10^0
55	5	0	1.85×10^{-1}	2.25×10^0
65	5	0	1.01×10^0	8.60×10^0
75	5	0	1.67×10^1	2.73×10^1
15	25	0	8.7×10^{-1}	Not investigated
25	25	0	1.75×10^0	Not investigated
35	25	0	3.35×10^0	Not investigated
45	25	0	7.56×10^0	Not investigated
55	25	0	3.64×10^1	Not investigated
35	0	5	2.39×10^{-4}	2.45×10^{-2}
45	0	5	1.5×10^{-3}	1.01×10^{-1}
55	0	5	1.14×10^{-2}	5.26×10^{-1}
65	0	5	2.59×10^{-1}	3.61×10^0
75	0	5	8.65×10^0	2.00×10^1
30	5	5	2.03×10^{-3}	Not investigated
40	5	5	9.60×10^{-3}	Not investigated
50	5	5	1.37×10^{-1}	Not investigated
60	5	5	1.02×10^0	Not investigated
70	5	5	1.30×10^1	Not investigated

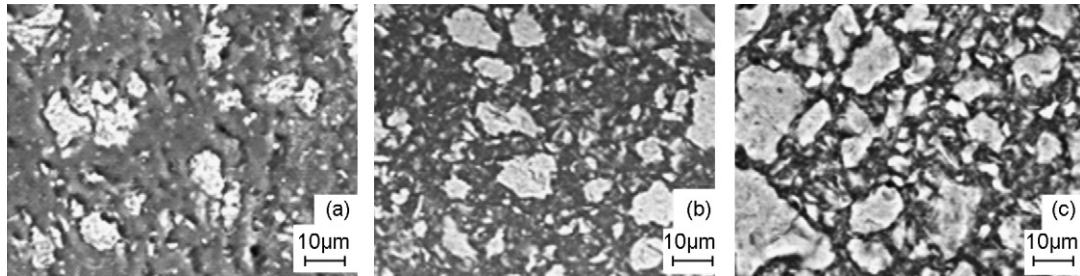


Fig. 2. SEM surface micrographs of PP/G at: (a) 40 wt.% G, (b) 60 wt.% and (c) 80 wt.%.

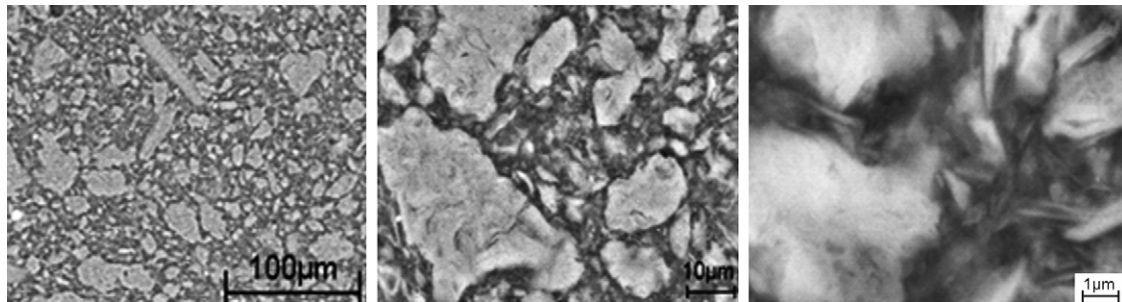


Fig. 3. SEM surface micrographs of PP/80 wt.% G at different magnifications.

concentration is increased further, the conductivity increases to 7 S cm^{-1} at 80 wt.% G. It is noticed that a higher concentration of G is needed to create a conductive path throughout the entire composite. This observation has also been reported by Thongruang et al. [36,37] who found that the threshold loading level of G to achieve measurable conductivity in HDPE was $\sim 50 \text{ wt.}\%$.

The scanning electron micrographs in Figs. 2–4 reveal that graphite primary particles aggregate into larger clusters with low aspect ratios. These tend to disperse more evenly into the polymer matrix, thereby resulting in fewer particle–particle contacts at a given loading level and, consequently, a lower conductivity polymer composite. In other words, the filler particles do not make contact with their nearest neighbours due to partial wetting by the polymer. Dawson and Adkins [38] proposed that the conductivity in G-filled polymers is regulated by inter-particle electron transfer at low filler loadings and by intra-particle electron transfer at high filler loadings.

It was difficult, however, to prepare well-dispersed PP/G composites with simple mixing techniques. The surface properties of the filler and polymer have a significant effect on the

conductivity of the composite by influencing the interaction between them. How well the polymer wets the surface of the filler can be quantified by the difference between the surface energies of the two materials. Smaller differences between the two surface energy values lead to better wetting of the filler by the polymer. Better wetting means that larger amounts of the polymer are coating the filler surface, which will alter the distribution of the filler within the matrix. This will increase the percolation threshold and the overall resistivity of the composite because larger amounts of the filler are required before the particles will come in to contact with each other. PP is hydrophobic and has relatively low surface free energy ($\sim 20\text{--}25 \text{ mJ/m}^2$) [39]. G is also strongly hydrophobic these may be a small difference between its surface energy and that of PP [40] which might lead to partial wetting and hence lower conductivity. This would explain why a somewhat larger difference between the surface energy of the filler and the polymer is desirable.

On the other hand, the solution blended PP/G composites have higher conductivities than the melt-compounded composites. It can be observed from Fig. 5 that the conductivity of

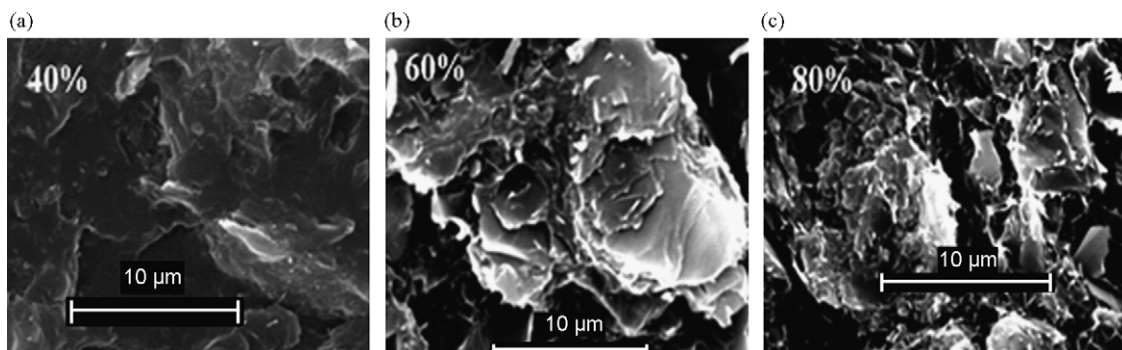


Fig. 4. SEM fracture micrographs of PP/G at: (a) 40 wt.% G, (b) 60 wt.% and (c) 80 wt.%.

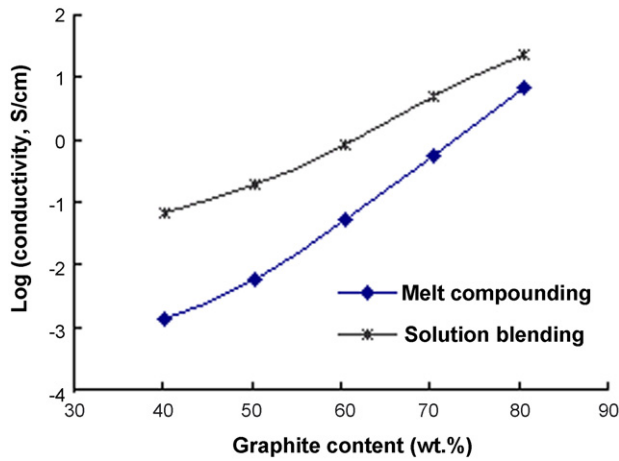


Fig. 5. Log conductivity vs. G content for melt compounding and solution blending of PP/G.

the composites increases from 7 S cm^{-1} for melt-compounded PP/G to 23 S cm^{-1} for solution-blended PP/G at 80 wt.% filler loading. The increase in conductivity may be attributed to the better filler distribution within the polymer matrix as reported by Radhakrishnan et al. [41] and Bennion et al. [42].

3.1.2. PP/G/CB composites

Composites containing a constant composition of PP (20 wt.%) and different compositions of G and CB have been investigated and their conductivities are plotted in Fig. 6 as a function of CB content. It is observed that the combination of G with CB has a significant effect on the conductivity. The maximum achievable conductivity is given by a combination of 25 wt.% CB and 55 wt.% G. It has a value that is five times higher than that of PP/G composites at the same filling degree (i.e., from 7 S cm^{-1} for PP/80%G to 36 S cm^{-1} for PP/G/CB). The conductivity of PP/G/CB starts to decrease on further additions of CB and this may be attributed to wetting limitations as reported by Mighri et al. [14], who observed that no more than 18 wt.% CB could be incorporated to PP/G composites due to such limitations. Moreover, it has been observed [43] that carbon blacks have better affinity for polar than for non-polar resins.

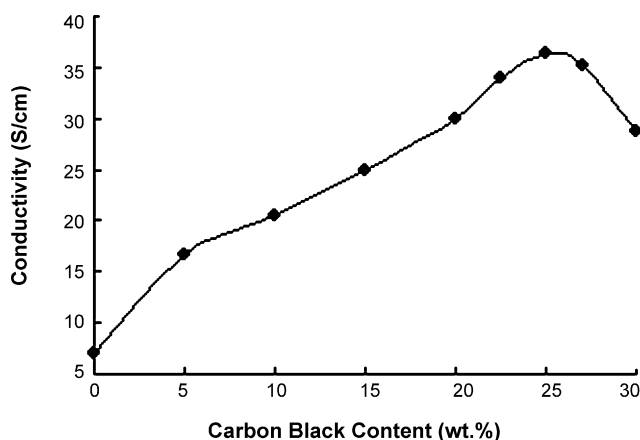


Fig. 6. Conductivity vs. CB content of PP/G/CB at 20 wt.% PP.

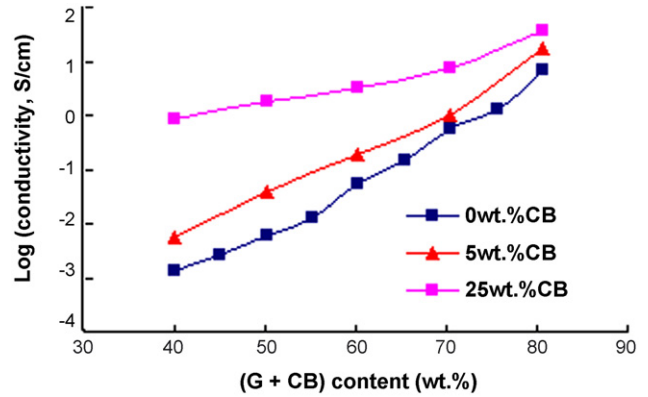


Fig. 7. Effect of CB on conductivity of PP/G/CB.

The effect of CB on the conductivities of the PP/G composites was further studied by varying the concentration of PP from 20 to 60 wt.% and CB from 5 to 25 wt.%, as shown in Fig. 7. The increase of conductivity is noticeable in all composites containing CB at all PP contents. Moreover, the difference in the conductivity of PP/G and PP/G/CB composites containing 25 wt.% CB is more pronounced at low rather than high filler (i.e., G and CB) concentrations. In other words, the $\sigma_{\text{PP/G/CB}}/\sigma_{\text{PP/G}}$ ratio is much larger for composites at 40 wt.% total filler loading than that at 80 wt.%. This difference becomes much smaller for composites containing 5 wt.% CB.

Solution blending of PP/G/CB composites was also investigated and gave better conductivities than that of melt compounding, as illustrated in Fig. 8. The content of CB in all composites was 5 wt.%. The conductivity increased from 16 to 27 S cm^{-1} by solution blending at 80 wt.% filling.

3.1.3. PP/G/PANi composites

The PANi was synthesized according to a method reported in the literature [34,35] without investigating in the effect of polymerization conditions on its conductivity. The polymerization conditions were set by the reaction temperature, oxidant

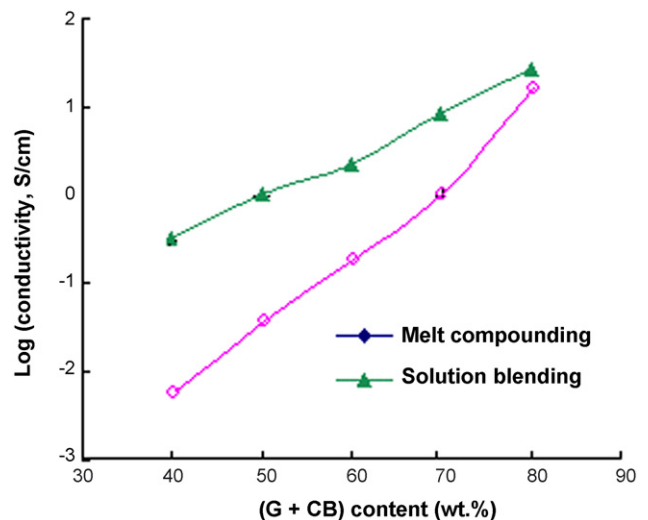


Fig. 8. Log conductivity vs. (G + CB) content for melt compounding and solution blending of PP/G/CB at 5 wt.% CB.

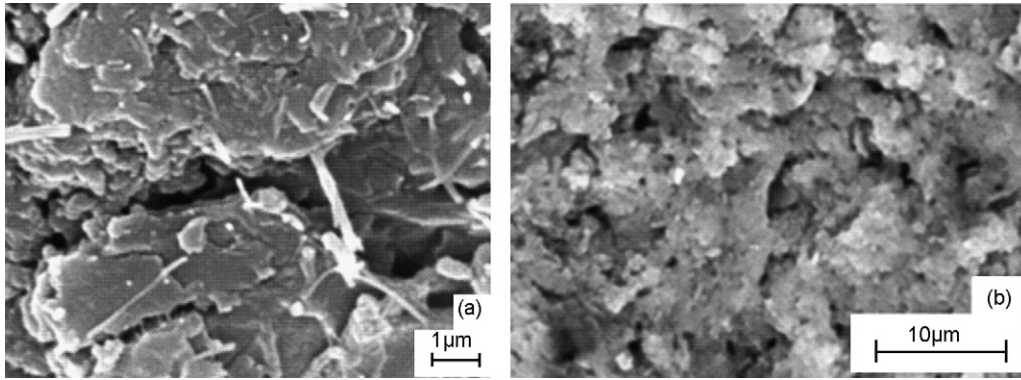


Fig. 9. SEM micrographs of PANi pellets (a) before doping with DBSA and (b) after doing with DBSA.

amount, additional HCl solution, and DBSA:aniline ratio. The resulting powders of PANi are homogeneous and form agglomerates with a solid rock shape and different sizes, as shown in Fig. 9(a). Doping PANi with DBSA results in small aggregate morphology, see Fig. 9(b).

The effect of PANi on the PP/G composites was first studied by incorporating 2 to 10 wt.% of PANi in different compositions of G at 20 wt.% PP. The data in Fig. 10 indicate a slight increase in the conductivity of the PP/G composite when including PANi up to 6 wt.%. This observation appears agree with that of Taipalus et al. [44], who found a positive influence of the PANi-complex on the electrical conductivity of long CF-PP composites. These synergistic effects were explained by enrichment of the PANi-complex at the interface between CF and PP. The conductivity of the composites decreases markedly when replacing G by more than 6 wt.% of PANi.

To study further the effect of PANi on the composites, 5 wt.% PANi was added to PP/G and PP/G/CB composites at different compositions of PP as illustrated in Fig. 11. Unlike the previous results, the conductivity of PP/G is decreased by incorporating 5 wt.% PANi at different compositions of PP. The same decrease in conductivity is also observed by adding PANi to PP/G/CB composites.

Solution-blended PP/G/PANi has higher conductivity than that of melt-compounded PP/G/PANi. This can be attributed to

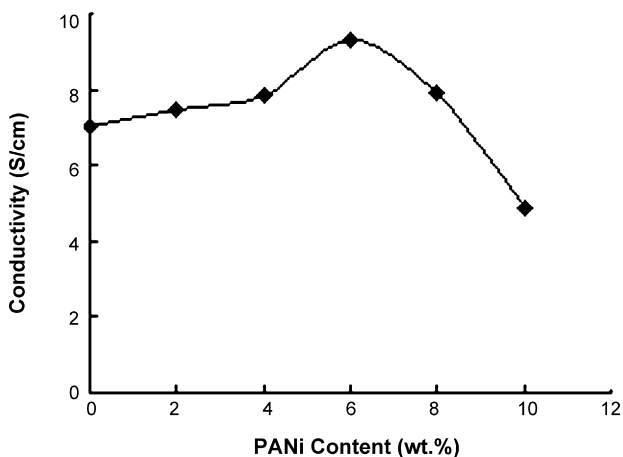


Fig. 10. Conductivity vs. PANi content of PP/G/PANi composites at 20 wt.% PP.

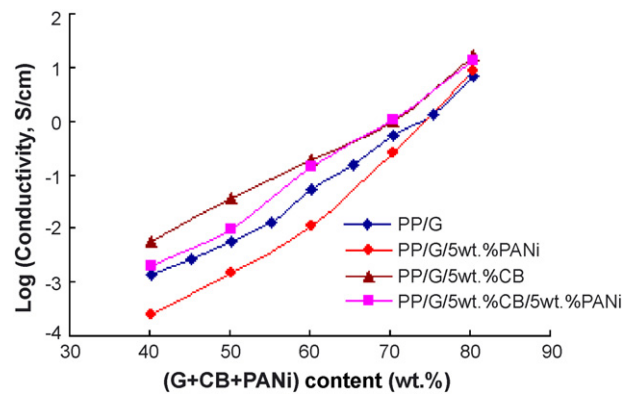


Fig. 11. Effect of PANi on conductivity of PP/G and PP/G/CB.

better distribution of graphite rather than to the addition of PANi. More results are shown in Fig. 12.

Lastly, a comparison of the conductivities for all the composites is presented in Fig. 13. It appears that the solution-blended composites lead to further improvement in electrical conductivity over that achieved by the melt-compounded process. The PP/G/CB composites have the highest conductivities while the PP/G/PANi composites showed the lowest conductivities.

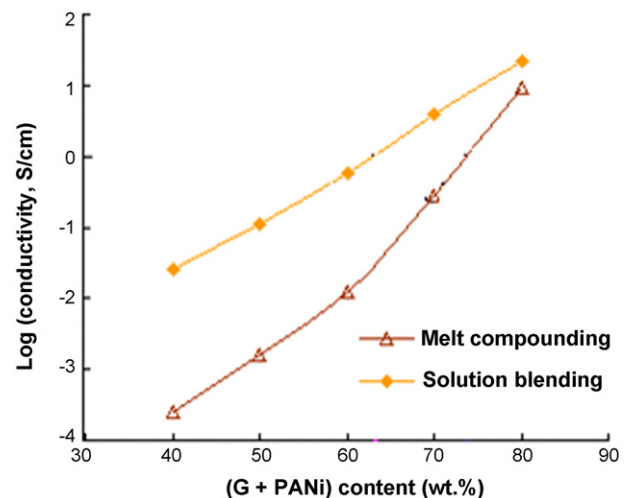


Fig. 12. Log conductivity vs. (G + PANi) content for melt compounding and solution blending of PP/G/PANi at 5 wt.% PANi.

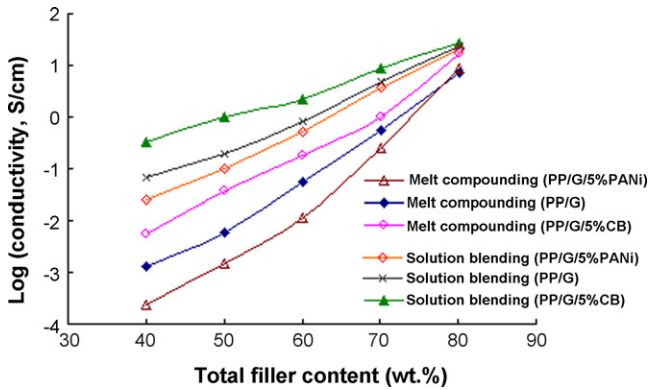


Fig. 13. Comparison of conductivities of melt-compounded and solution-blended PP/G, PP/G/CB, and PP/G/PANi composites.

The decrease in conductivity of composites containing PANi requires further investigation.

3.2. Thermogravimetric analysis of PANi and PANi composites

Thermogravimetric analysis (TGA) is a useful technique for characterizing the thermal stabilities of conducting polyaniline

composites. Thermogravimetric (TG) and derivative thermogravimetric (DTG) plots for G, PANi, PP/G and PP/G/PANi are given in Fig. 14. It is found that PANi undergoes three weight loss processes. The first loss, located below 110 °C, is attributed to the evaporation of residual moisture and oligomers. The second weight loss at 110–210 °C can be related to the decomposition of the DBSA dopant. The third and the final weight loss at 210–495 °C can be attributed to the degradation of the PANi backbones [45]. The total mass loss for PANi was 64.2%.

The derivative thermogravimetric curve for PANi has three peaks, namely, a minor one at 50 °C and two major ones at 170 and 280 °C. For PP/G with a 60 wt.% G content, degradation is observed in a single step up to 480 °C. Thereafter, a sharp weight loss occurs in the material. For PP/G/PANi at 70 wt.% G and 10 wt.% PANi, minor peaks occur before the major one at 465 °C.

A step analysis of the composites was performed. The weight losses at different temperatures are given in Table 2.

The PANi has unsuitably poor thermal stability to undergo melt compounding. As a matter of fact, useful commercial applications of PANi require a fine balance of conductivity, processability and stability. Until recently, however, material scientists have been unable to obtain all three properties simul-

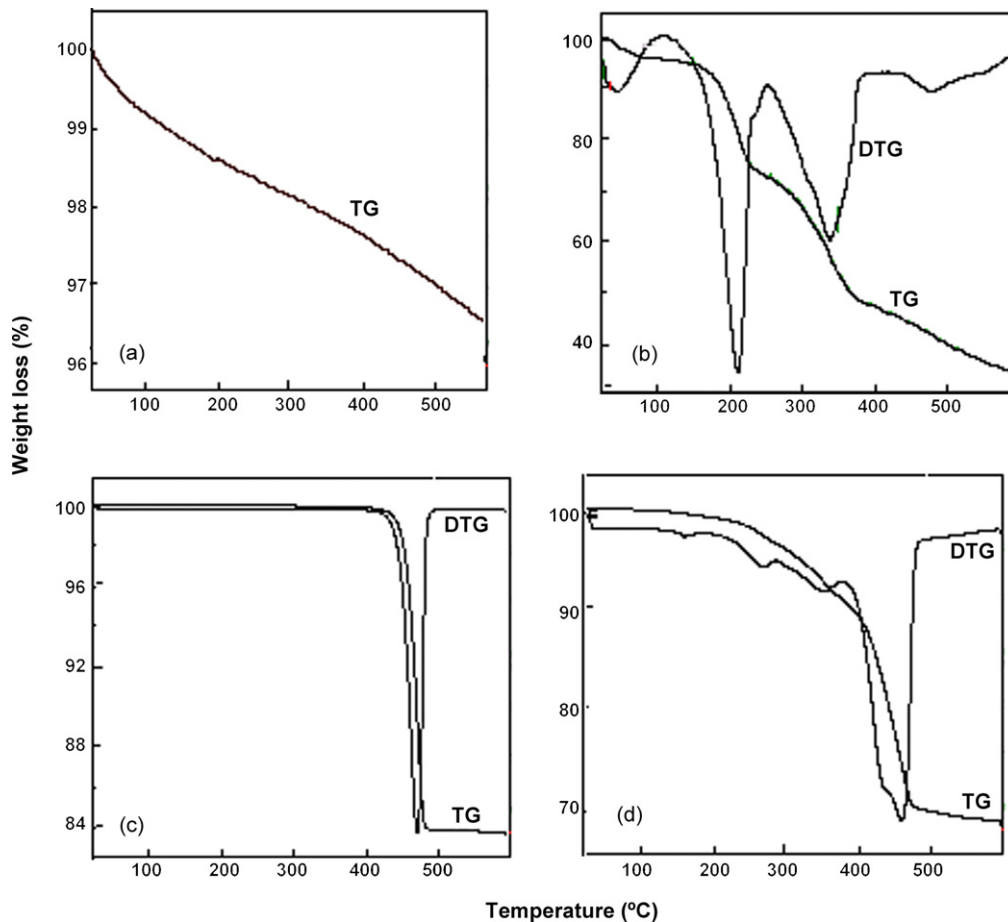


Fig. 14. TG and DTG of: (a) G, (b) PANi, (c) PP/G and (d) PP/G/PANi composites.

Table 2
Percentage weight losses of composites at different temperatures

Sample	Weight loss (%)								
	50 °C	100 °C	150 °C	200 °C	250 °C	300 °C	400 °C	500 °C	600 °C
G	0.4	0.9	1.3	1.5	1.8	1.9	2.4	3.0	3.5
PANi	2.0	4.0	5.1	12.5	25.6	31.4	50.1	57.8	64.2
PP/G	0.0	0.04	0.09	0.09	0.09	0.09	0.09	16.3	16.3
PP/G/PANi	0.1	0.20	0.40	0.90	1.80	4.00	10.5	30.1	31.1

taneously and that might limit the use of ICPs with CB or G as conductive additives in the thermoplastics industry, in addition to the undesirable properties of these materials at high temperatures. The conductivity in PANi-complex blended with thermoplastics depends on many factors such as the fraction of PANi-complex, the processing temperature, the shear stress applied, the viscosity of the matrix polymer, and the surface activity of the PANi-complex.

4. Conclusions

The electrical properties of PP/G composites have been investigated in an attempt to understand the mechanism of conduction in these composites for application in bipolar plates in PEM fuel cell. The conductivity of PP/G is relatively low and to improve it, a homogenous dispersion of the graphite in PP must occur. This is difficult, however, to obtain by a melt-compounding process. Combining G with CB is an effective way to develop higher conductivity composites. The best conductivity results for the bipolar plate are obtained by combining 25 wt.% CB and 55 wt.% G with 20 wt.% PP. Solution blending leads to further improvement in electrical conductivity compared with melt compounding. Finally, investigations reveal that composites containing PANi have lower conductivity. This is attributed to the poor thermal stability of PANi to undergo melt compounding.

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References

- [1] V. Metha, J.S. Cooper, *J. Power Sources* 5044 (2002) 1–22.
- [2] M.H. Oh, Y.S. Yoon, S.G. Park, *Electrochim. Acta* 50 (2004) 777–780.
- [3] J. Huang, D.G. Baird, J.E. McGrath, *J. Power Sources* 150 (2005) 110–119.
- [4] M.S. Wilson, C. Zawodzinski, G. Bender, T.A. Zawodzinski, D.N. Busick, *Proceedings of the 2000 DOE Hydrogen Program Review*, 2000.
- [5] E. Balko, N. Edward, R. Lawrence, J. Richard, US patent 4,339,322 (1982).
- [6] M.S. Wilson, S. Mahlon, D.N. Busick, N. Deanna, US patent 6,248,467 (2001).
- [7] T.M. Besmann, J.W. Klett, T.D. Burchell, *Mater. Res. Soc. Symp. Proc.* 496 (1998) 243.
- [8] K. Ledjeff-Hey, F. Mahlendorf, O. Niemzing, A. Trautmann, *Fuel Cell Seminar*, Palm Spring, CA, November 1998, 1998.
- [9] A. Heinzl, F. Mahlendorf, O. Neimzig, C. Kreuz, *J. Power Sources* 131 (2004) 35–40.
- [10] E.A. Cho, U.-S. Jeon, H.Y. Ha, S.-A. Hong, I.-H. Oh, *J. Power Sources* 125 (2004) 178–182.
- [11] H.-C. Kuan, C.-C.M. Ma, K.H. Chen, S.-M. Chen, *J. Power Sources* 134 (2004) 7–17.
- [12] E. Middelma, W. Kout, B. Vogelaar, J. Lenssen, E. de Waal, *J. Power Sources* 118 (2003) 44–46.
- [13] Product Information, DuPont Fuel Cells, Conductive Bipolar Plates, T8 Series.
- [14] F. Mighri, M.A. Huneault, M.F. Champagne, *Poly. Eng. Sci.* 44 (2004) 1755–1765.
- [15] Y. Wang, *Conductive Thermoplastic Composite Blends for Flow Field Plates for Use in Polymer Electrolyte Membrane Fuel Cell (PEMFC)*, M.S. Thesis., University of Waterloo, Ontario, Canada, 2006.
- [16] W. Vielstich, H.A. Gasteiger, A. Lamm, *Handbook of Fuel Cells—Fundamentals, Technology and Applications*, vol. 3, John Wiley & Sons, Ltd, 2003, 274–313.
- [17] V. Causin, C. Marega, A. Marigo, G. Ferrara, A. Ferraro, *Eur. Poly. J.* 42 (2006) 3153–3161.
- [18] C. Del Rio, M.C. Ojeda, J.L. Acosta, M.J. Escudero, E. Hontanon, L. Daza, *J. Appl. Poly. Sci.* (2002) 2817–2822.
- [19] R. Kaiser, H.-G. Fritz, *The Polymer Processing Society, 17th Annual Meeting*, Montreal, Canada, May 21–24, 2001.
- [20] J.F. Yacubowicz, M. Narkis, *Polym. Eng. Sci.* 30 (1990) 459–468.
- [21] G.N. Lee, K.D. Suh, *Polym. Eng. Sci.* 38 (1998) 471–477.
- [22] B.-L. Lee, *Polym. Eng. Sci.* 32 (1992) 36.
- [23] I. Novak, I. Krupa, I. Chodak, *Eur. Poly. J.* 39 (2003) 585–592.
- [24] W. Li, C.L. Johnson, H.L. Wang, *J. Appl. Polym. Sci.* 51 (1994) 1347–1349.
- [25] S.W. Byun, S.S. Im, *Synth. Met.* 69 (1995) 219–220.
- [26] M. Špírková, J. Stejskal, O. Quadrat, *Synth. Met.* 102 (1999) 1264–1265.
- [27] M. Chipara, D. Hui, P.V. Notinger, M.D. Chipara, K.T. Lau, J. Sankar, D. Panaitescu, *Compos. Part B* 34 (2003) 637–645.
- [28] R.K. Gupta, R.A. Singh, *J. Non-Cryst. Solids* 351 (2005) 2022–2028.
- [29] P.L.B. Araújo, E.S. Araújo, R.F.S. Santos, A.P.L. Pacheco, *Microelectron. J.* 36 (2005) 1055–1057.
- [30] J. Anand, S. Palaniappanb, D.N. Sathyanarayana, *Prog. Poly. Sci.* 23 (1998) 993–1018.
- [31] A. Bhattacharya, A. De, *Prog. Solid State Chem.* 24 (1996) 141–181.
- [32] A. Pud, N. Ogurtsova, A. Korzhenkob, G. Shapovalva, *Prog. Poly. Sci.* 28 (2003) 1701–1753.
- [33] S. Joseph, J.C. McClure, R. Chianelli, P. Pich, P.J. Sebastian, *Int. J. Hydro. Energ.* 30 (2005) 1339–1344.
- [34] W. Yin, E. Ruckenstein, *Synth. Met.* 108 (2000) 39–46.
- [35] S.J. Su, N. Kuramoto, *Synth. Met.* 108 (2000) 121–126.
- [36] W. Thongruang, R.J. Spontak, C.M. Balik, *Polym.* 43 (2002) 2279–2286.
- [37] W. Thongruang, R.J. Spontak, C.M. Balik, *Polym.* 43 (2002) 3717–3725.
- [38] J.C. Dawson, C.J. Adkins, *J. Phys. Condens. Mat.* 8 (1996) 8321.
- [39] N. Inagak, *Plasma surface Modification and Plasma Polymerization*, Technomic Publication Company, Lancaster, 1996.

- [40] M.L. Clingerman, Development and Modeling of Electrically Conductive Composite Materials, PhD Thesis., Michigan Technological University, England, 2001.
- [41] S. Radhakrishnan, B.T.S. Ramanujam, A. Adhikari, S. Sivaram, *J. Power Sources* 163 (2007) 702–707.
- [42] Bennion, N. Douglas, Lafollette, M. Rodney, Stewart, L. Lance, US patent 5,141,828, August 25 (1982).
- [43] V. Haddadi-Asl, M. Kazacos, M. Skyllas-Kazacos, *J. Appl. Electrochem.* 25 (1995) 19.
- [44] R. Taipalus, T. Harmia, M.Q. Zahang, K. Friedrich, *Compos. Sci. Tech.* 61 (2001) 801–814.
- [45] D.T.C. Castro, M.M.C. Ortega, I. Villarreal, H. Grijalva, M.P.T. Tello, S.M.N. Donlucas, J.E.L. Puig, *Compos.: Part A* 37 (2006) 5493–5499.