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# Effect of Moisture and Temperature on Mechanical Properties of Graphite Composite Bipolar Plate for Proton Exchange Membrane Fuel Cell (PEMFC)

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#### Abstract

The fuel cell is considered to be a promising alternative energy source for the near future. A bipolar plate, which is the major component of the fuel cell, is always exposed to the moisture during operation, because one of the important functions of the bipolar plate is to facilitate water management within the cell. In order to use graphite composite materials as bipolar plate material for PEMFC, the effect of the moisture environment on the mechanical properties of bipolar plate is studied. Two types of specimens, one made of particulate graphite 90%-epoxy 10% composite 'with' woven carbon fabric, and the other 'without' carbon fabric, were fabricated. Both types of specimens were kept in three different environmental conditions: (1) dry at room temperature, (2) immersed in 85°C water for 100 h, and (3) immersed in 85°C water for 300 h. A series of experiments were performed with the samples of the two types: water absorption rate, strength and modulus from the bending and tensile tests were measured. The results showed that a steep increase of water absorption rate occurred at the beginning of the test followed by a slow increase afterwards. Total water absorption rate was lower for the specimens with fabric since the inserted carbon fabric has lower porosity and lower water absorption than the specimens without fabric made of graphite/epoxy composite. Bending strength and modulus decreased for both types of specimens. However, addition of carbon fabrics to the graphite-particle/epoxy composite increased the tensile strength significantly. © Koninklijke Brill NV, Leiden, 2011

#### Keywords

Fuel cell, bipolar plate, environmental degradation, mechanical testing

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### 1. Introduction

The proton exchange membrane fuel cell (PEMFC) is considered to be a promising alternative energy source for the near future. PEM fuel cells are favored especially for use in automobiles due to the high power density, relatively quick start-up, rapid response to varying loads, and low operating temperature [1–3]. The bipolar plate, which is the major component of the fuel cell, has important functions in the PEM fuel cell. The bipolar plate distributes the fuel and oxidant within the cell, facilitates water management within the cell, separates the individual cells in the stack, carries current away from the cell, supports thin membrane and withstands the clamping forces of the stack assembly [3]. Clamping force is a critical factor influencing the performance of the membrane electrode assembly (MEA). If the clamping pressure is too small, the interfacial contact resistance between the MEA and the bipolar plate will increase, decreasing the efficiency of the fuel cell system. The efficiency also decreases if the clamping pressure is too large [26]. For these reasons, bipolar plates should possess high enough mechanical strength and stiffness to support thin membranes and electrodes, and to withstand the high clamping forces in the stack assembly. Since the bipolar plate plays such important functions, various studies on the bipolar plate have been performed.

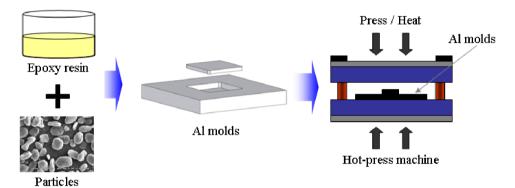
Mehta and Cooper provided a comprehensive review of design and manufacturing alternatives for bipolar plates [3]. Heinzel *et al*. [4] and Heo *et al*. [5] introduced new manufacturing methods of bipolar plates individually. New materials for bipolar plates have been proposed by many researchers. Wind *et al*. [6] and Hodgson *et al*. [7] demonstrated coated metallic plates for the bipolar plate. Kumar and Reddy [8] proposed Ni–Cr metal foam as an alternative material for the bipolar plate. Due to the chemical instability of the metals in the fuel cell environments, corrosion of metal bipolar plates has been discussed by various researchers [9–11]. Composite materials were also investigated extensively for bipolar plate application, and Wu and Shaw studied functional improvement of polymer bipolar plates by adding nanotubes [12]. Electrical, thermal and mechanical properties of composite bipolar plates have been evaluated by several studies [13–18].

PEM fuel cells are operated at temperatures higher than 80°C and under high humidity [28]. Since the properties of composite bipolar plates could be affected by the operating environment, the effect of the environment on properties must also be understood. While the effects of humidity and temperature on the mechanical properties of proton exchange membrane have been studied [19, 28], there are insufficient investigations to understand the effect of the environment on the properties of the composite bipolar plate.

In this study, the bipolar plate samples were constructed and tested to investigate the effect of the moisture and temperature environment on the mechanical properties of the graphite particle/epoxy composite as a material for bipolar plate. Also, the effect of incorporating carbon woven fabric on mechanical properties was investigated.

Material Epoxy resin (YD-128)	Property	
	Density Viscosity	1.17 g/cc 11 500–13 500 cps at 25°C
Graphite particles (P-15)	Density Diameter	0.9 g/cc 18.8 µm
Carbon fabric (KN C123 EPC)	Thickness Mass per area	160 $\mu$ m/ply 290 $\pm$ 15 g/ $m^2$

**Table 1.**Properties of epoxy resin, graphite particle and carbon fabric



**Figure 1.** Manufacturing progress of carbon composite. This figure is published in color on http://www.ingentaconnect.com/content/vsp/acm

#### 2. Environmental

## 2.1. Preparation of Test Specimens

The material used in this research is shown in Table 1. The graphite particle and resin was processed in proportions of 9:1 by volume. Manufacturing process of carbon composite is shown in Fig. 1. First, top and bottom of aluminum molds were fabricated by machining. Second, particles were mixed with epoxy resin, and the mixture was spread over the lower mold. Third, the mold was closed and pressure (10 MPa) and temperature (120°C) were applied. Finally, the mold must be cooled to solidify the mixture, and specimen was taken out from the mold [20]. Two types of bipolar plate specimens, one made of particulate graphite—epoxy composite with carbon fabric and the other without carbon fabric, were fabricated (Fig. 2). Here, the carbon fabric was inserted in the middle of the bipolar plate layer in order to improve mechanical properties.

# 2.2. Experimental Procedure

An environmental test was conducted to determine tensile strength, modulus and bending strength, modulus and water absorption rate. The mechanical properties of

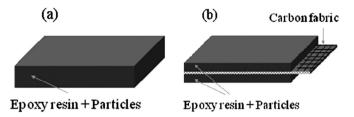


Figure 2. Geometry of specimens for environmental test: (a) without fabric and (b) with fabric.

**Table 2.** Test matrix of environmental test

Environmental conditions	Type of specimens
Dry/room temp. (25°C)	Without fabric (graphite/epoxy) With fabric (graphite/epoxy/fabric)
Water/85°C temp. (100 h)	Without fabric (graphite/epoxy) With fabric (graphite/epoxy/fabric)
Water/85°C temp. (300 h)	Without fabric (graphite/epoxy) With fabric (graphite/epoxy/fabric)

<sup>\*</sup>For each type, five specimens were tested.

composite have been investigated at different humidity and temperature in an environmental chamber (Daihan-Brand, DWTH-155). Table 2 shows the environmental conditions as a test matrix.

# 2.2.1. Water Absorption Rate

Specimens were placed into a distilled water chamber at 85°C. Once the specimens were taken out from the water chamber, water drops at the surface was absorbed by dry paper towel. Then specimens were weighed by using a precision balance with 0.001 g resolution, and the percentage in weight change was determined. The weight gain was taken as the moisture absorption, using the following formula [21]:

$$M_t = \frac{m_t - m_0}{m_t} \times 100, (1)$$

where  $m_0$  is the initial mass of the specimen,  $m_t$  is the mass of the specimen after immersion for time t in hours, and  $M_t$  is the percent moisture content of the specimen.

The water diffusivity of the composite material, D, was obtained from the expression

$$D_{\rm a} = \pi \left(\frac{h}{4M_{\rm m}}\right)^2 \left(\frac{M_2 - M_1}{\sqrt{t_2} - \sqrt{t_1}}\right)^2,\tag{2}$$

where  $M_{\rm m}$  is the maximum moisture content (weight gain), and h is the specimen thickness (h=4 mm).  $M_1$  and  $M_2$  are the moisture contents at time  $t_1$  and  $t_2$ ,

respectively. Because of the dimensions of the specimen used, the application of an edge correction was necessary and the actual sample diffusivity could be determined as

$$D_{x} = D_{a} \left( 1 + \frac{h}{l} + \frac{h}{w} \right)^{-2}, \tag{3}$$

where l and w are the sample length (l=80 mm) and width (w=10 mm), respectively [33]. The diffusivity of the graphite composite with fabric ( $M_{\rm m}=1.888\%$ ) and without fabric ( $M_{\rm m}=2.850\%$ ) are  $1.90\times10^{-5}$  mm<sup>2</sup>/s and  $2.93\times10^{-5}$  mm<sup>2</sup>/s, respectively. The change in the moisture content as a function of time could be expressed as the following equation [22, 23]:

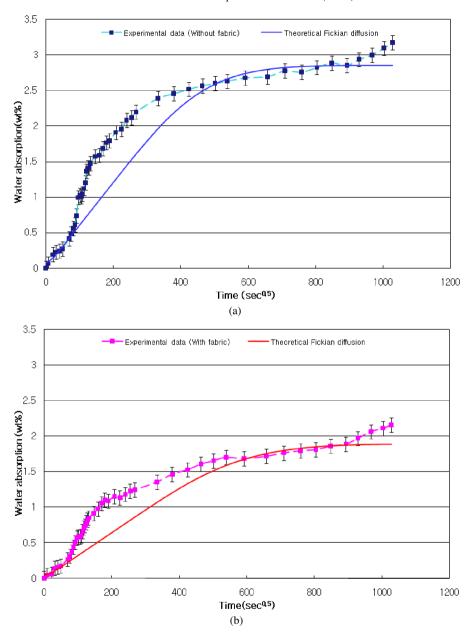
$$M = M_{\rm m} \left( 1 - \frac{8}{\pi^2} \sum_{j=0}^{\infty} \frac{\exp[-(2j+1)^2 \pi^2 (Dt/h^2)]}{(2j+1)^2} \right). \tag{4}$$

In Fig. 3, the weight change profiles are demonstrated for the two types of specimens, one with fabric and the other without fabric. The result of the test revealed that both specimens made with graphite composite, with fabric and without fabric, showed high water absorption at the beginning of the test. The absorption rate of the graphite composite without fabric was about 1.5 times higher than that of the graphite composite with fabric.

The specimen's data diverge from Fickian behavior before reaching the saturation level, as the experimental weight change is smaller than that of the theoretical values. The change in moisture content in both types of specimens could be expressed using typical hygrothermal theory of a graphite/epoxy composite. Imaz et al. [24] found that the moisture absorption of graphite/epoxy composites was Fickian, but at higher temperature, there is a low correlation for the saturation level to increase due to material degradation. This trend could be attributed to a slight irreversible degradation in the material. Loos and Springer [25] investigated the moisture absorption behavior of many kinds of resin-matrix composites. Their results show that materials obey Fickian diffusion behavior at lower temperatures and non-Fickian behavior at higher temperatures. The plausible explanation is due to the moist, high temperature environment, and microcracks developed on the surface and inside the materials. As the cracks developed, material was actually lost, most likely in the form of resin particles. As long as the moisture gain was greater than the material loss, the weight of the specimen increased. However, for the composite bipolar plates, Du and Jana [28] reported that the moisture absorption followed linear Fickian diffusion behavior after 6 months.

# 2.2.2. Bending Strength and Modulus

A bipolar plate requires mechanical strength to withstand clamping forces of the fuel cell package. The three-point bending tests were conducted to determine flexural modulus and strength using with Lloyd Instruments LR 50K according to ASTM D 790. The dimensions of the specimen are shown in Fig. 4.



**Figure 3.** Water absorption rate according to circumstance condition: (a) without fabric and (b) with fabric. This figure is published in color on http://www.ingentaconnect.com/content/vsp/acm

Figure 5 shows failure modes of the bending specimens, and Fig. 6 shows the results of bending test. The flexural strengths of 'with fabric' and 'without fabric' specimen were higher than the DOE target value, 34 MPa. However, specimens with fabric generally show lower flexural strength and modulus than specimens without



Figure 4. Dimensions of bending specimen (ASTM D 790).

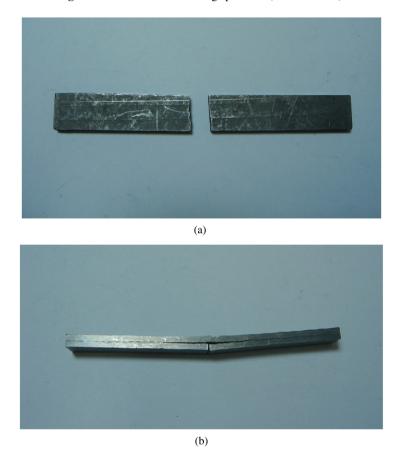
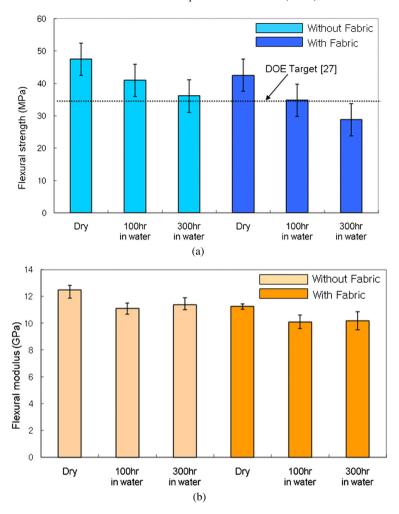


Figure 5. Failure modes of bending specimens: (a) without fabric (top view) and (b) with fabric (side view).

fabric, and the fractured specimens with fabric showed that only one side of the specimens was broken. In the case of the specimens without fabric, the specimens immersed in 85°C for 300 h showed flexural strength lower by 24%, and by 10% in flexural modulus than the specimen tested dry and at room temperature condition. The graphite composite with fabric, with the specimens immersed at 85°C for 300 h, showed flexural strength lower by 34%, and by 10% in flexural modulus compared to the specimens tested dry and at room temperature condition.



**Figure 6.** Results of the bending tests: (a) strength and (b) modulus. This figure is published in color on http://www.ingentaconnect.com/content/vsp/acm

## 2.2.3. Tensile Strength and Modulus

Tensile tests were performed with Lloyd Instruments LR 50K according to ASTM D 638. The dimensions of the tensile specimen are shown in Fig. 7. Figure 8 shows the failure modes of the tensile specimens, and Fig. 9 shows the results of the tensile test. The tensile strength of 'with fabric' specimen was higher than those of several polymer composite bipolar plates reported by other researchers [29–32]. Also, tensile strength and modulus increased by 46–66% and 20–39%, respectively, when carbon fabric was added in the composite. Without carbon fabric, the composite was brittle due to the small content of epoxy resin that played the role of binder in the composite. However, with fabric, the composites were able to delay brittle fracture by the aid of reinforcement. In the case of the graphite composite without

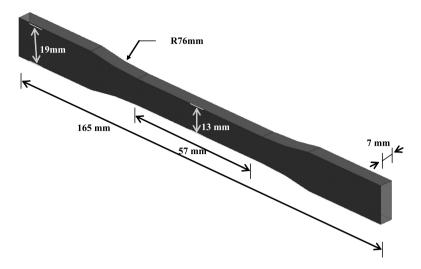
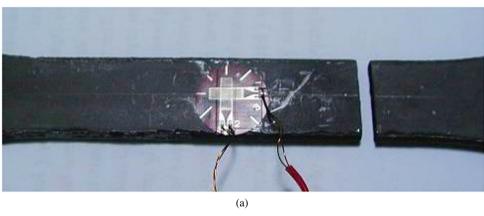
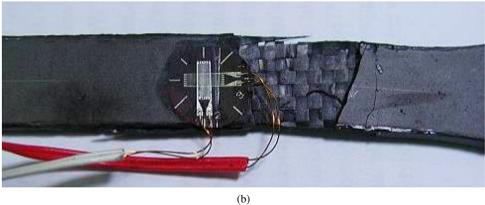
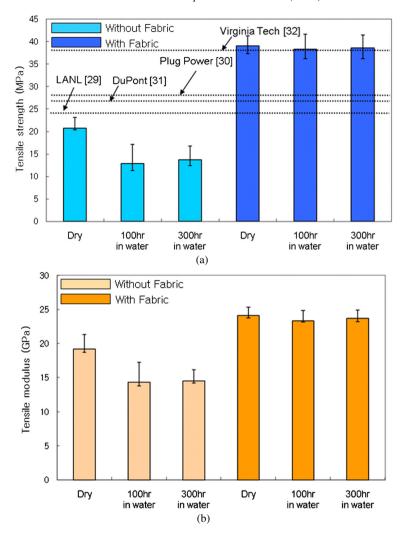


Figure 7. Dimensions of tensile specimen (ASTM D 638).





**Figure 8.** Failure modes of tensile specimens: (a) without fabric and (b) with fabric. This figure is published in color on http://www.ingentaconnect.com/content/vsp/acm



**Figure 9.** Results of the tensile test: (a) strength and (b) modulus. This figure is published in color on http://www.ingentaconnect.com/content/vsp/acm

fabric, the specimens immersed in 85°C for 300 h showed tensile strength lower by 35%, and by 24% in tensile modulus than the specimens tested dry and in room temperature conditions. The graphite composite with fabric, the specimens immersed in 85°C for 300 h, showed tensile strength and modulus almost the same as the specimen tested at dry and room temperature condition.

### 3. Conclusions

The effect of water absorption on the mechanical properties of two types of graphite composite for the PEMFC bipolar plate has been studied. Experimental results show that addition of carbon fabric to the graphite-particle/epoxy decreased water absorp-

tion of the bipolar plate. Tensile strength and modulus decreased for the composites 'without fabric' in the hot water condition, but these values remained almost the same for the specimens 'with fabric'. The flexural strengths of 'with fabric' and 'without fabric' specimen were higher than the target DOE target value of 34 MPa. However, specimens 'with fabric' relatively showed a little lower flexural strength and modulus. Thus, moisture and high temperature reduced tensile and bending strength, but the addition of carbon fabrics to the graphite-particle/epoxy composite increased tensile strength significantly.

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### References

- S. Gottesfeld, T. A. Zawodzinski, T. E. Springer, M. S. Wilson and C. Zawodzinski, PEM fuel cells for transportation and stationary power generation applications, *Intl. J. Hydrogen Energy* 22, 1137–1144 (1997).
- 2. EG & G Services, Parsons Inc. and Science Applications International Corporation, *Fuel Cell Handbook*, 5th edn., U.S. Department of Energy, Morgantown, WV (2000), available at http://www.fuelcells.org/info/library/fchandbook
- V. Mehta and J. S. Cooper, Review and analysis of PEM fuel cell design and manufacturing, J. Power Sources 114, 32–53 (2003).
- 4. A. Heinzel, F. Mahlendorf, O. Niemzig and C. Kreuz, Injection moulded low cost bipolar plates for PEM fuel cells, *J. Power Sources* **131**, 35–40 (2004).
- S. I. Heo, K. S. Oh, J. H. Jang, S. H. Jung, Y. C. Yang and K. S. Han, A study on procuring condition of the 2-step manufacturing method for PEMFC composite bipolar plates, *J. Korean Soc. Compos. Mater.* 20, 10–16 (2007).
- J. Wind, R. Spah, W. Kaiser and G. Bohm, Metallic bipolar plates for PEM fuel cells, *J. Power Sources* 105, 256–260 (2002).
- D. R. Hodgson, B. May, P. L. Adcock and D. P. Davies, New lightweight bipolar plate system for polymer electrolyte membrane fuel cells, *J. Power Sources* 96, 233–235 (2001).
- A. Kumar and R. G. Reddy, Materials and design development for bipolar/end plates in fuel cells, J. Power Sources 129, 62–67 (2004).
- S. A. Gmboa, J. G. Gonzalez-Rodriguez, E. Valenzuela, B. Campillo, P. J. Sebastian and A. Reyes-Rojas, Evaluation of the corrosion resistance of Ni–Co–B coatings in simulated PEMFC environment, *Electrochimica Acta* 51, 4045–4051 (2006).
- M. C. Li, C. L. Zeng, S. Z. Luo, J. N. Shen, H. C. Lin and C. N. Cao, Electrochemical corrosion characteristics of type 316 stainless steel in simulated anode environment for PEMFC, *Electrochimica Acta* 48, 1735–1741 (2003).
- 11. M. Li, S. Luo, C. Zeng, J. Shen, H. Lin and C. Cao, Corrosion behavior of TiN coated type 316 stainless steel in simulated PEMFC environments, *Corrosion Science* **46**, 1369–1380 (2004).

- 12. M. Wu and L. L. Shaw, A novel concept of carbon-filled polymer blends for applications in PEM fuel cell bipolar plates, *Intl J. Hydrogen Energy* **30**, 373–380 (2005).
- E. A. Cho, U. S. Jeon, H. Y. Ha, S. A. Hong and I. H. Oh, Characteristics of composite bipolar plates for polymer electrolyte membrane fuel cells, *J. Power Sources* 125, 178–182 (2004).
- 14. H. C. Kuan, C.-C. M. Ma, K. H. Chen and S. M. Chen, Preparation, electrical, mechanical and thermal properties of composite bipolar plate for a fuel cell, *J. Power Sources* **134**, 7–17 (2004).
- A. Yasmin and I. M. Daniel, Mechanical and thermal properties of graphite platelet/epoxy composites, *Polymer* 45, 8211–8219 (2004).
- M. H. Oh, Y. S. Yoon and S. G. Park, The electrical and physical properties of alternative material bipolar plate for PEM fuel cell system, *Electrochimica Acta* 50, 777–780 (2004).
- 17. M. K. Seo and S. J. Park, Electrical resistivity and rheological behaviors of carbon nanotubes-filled polypropylene composites, *Chem. Phys. Letts* **395**, 44–48 (2004).
- 18. I. Krupa, I. Novak and I. Chodak, Electrically and thermally conductive polyethylene/graphite composites and their mechanical properties, *Synthetic Metals* **145**, 245–252 (2004).
- Y. Tang, A. M. Karlsson, M. H. Santare, M. Gilbert, S. Cleghorn and W. B. Johnson, An experimental investigation of humidity and temperature effects on the mechanical properties of perfluorosulfonic acid membrane, *Mater. Sci. Engng: A* 425, 297–304 (2006).
- H. S. Lee, H. J. Kim, S. G. Kim and S. H. Ahn, Evaluation of graphite composite bipolar plate for PEM (proton exchange membrane) fuel cell: electrical, mechanical, and molding properties, *J. Mater. Proc. Technol.* 187–188, 425–428 (2007).
- 21. C. H. Shen and G. S. Springer, Moisture absorption and desorption of composite materials, *Compos. Mater.* **10**, 2–20 (1976).
- R. F. Gibson and F. Ronald, *Principles of Composite Material Mechanics*, 2nd edn. McGraw-Hill Inc., NY (1994).
- S. W. Tsai and H. T. Hahn, Introduction to Composite Materials. Technomic Publishing, Lancaster, PA (1980).
- J. J. Imaz, J. L. Rodriguez, A. Rubio and I. Mondragon, Hydrothermal environment influence on water diffusion and mechanical behavior of carbon fiber/epoxy laminates, *J. Mater. Sci. Letts* 10, 662–665 (1991).
- 25. A. C. Loos and G. S. Springer, Moisture absorption of graphite-epoxy composition immersed in liquids and humid air, in: *Environmental Effects on Composite Materials*, G. Springer (Ed.), pp. 34–50. Technomic Publishing Company, Westport, CT (1981).
- P. Zhou, C. W. Wu and G. J. Ma, Contact resistance prediction and structure optimization of bipolar plate, *J. Power Sources* 159, 1115–1122 (2006).
- 27. Washington, DC, US Department of Energy, *The Hydrogen, Fuel Cells and Infrastructure Technologies Program Multi-Year Research*, pp. 3–89 (2005).
- L. Du and S. C. Jana, Hygrothermal effects on properties of highly conductive epoxy/graphite composites for applications as bipolar plates, J. Power Sources 182, 223–229 (2008).
- 29. M. S. Wilson and D. N. Busick, US Patent 6,248,467 (2001).
- J. G. Clulow, F. E. Zappitelli, C. M. Carlstrom, J. I. L. Zemsky, D. N. Busick and M. S. Wilson, AIChE Spring National Meeting, March 10–14, New Orleans, LA, pp. 417–425 (2002).
- 31. http://www.dupont.com/fuelcells/pdf/plates.pdf
- 32. J. Huang, D. G. Baird and J. E. McGrath, Development of fuel cell bipolar plates from graphite filled wet-lay thermoplastic composite materials, *J. Power Sources* **150**, 110–119 (2005).
- 33. L. R. Bao and A. F. Yee, Effect of temperature on moisture absorption in a bismaleimide resin and its carbon fiber composites, *Polymer* **43**, 3987–3997 (2002).