

New Approach to Fabricate Densificated Continuous Fiber Reinforced Polytetrafluoroethylene Composites with Significantly Improved Interfacial Bonding and Mechanical Properties

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ABSTRACT: An unique two-step technique to fabricate densified continuous fiber reinforced polytetrafluoroethylene (PTFE) composites with significantly improved interfacial bonding and mechanical properties have been successfully developed. The technique is conventional sintering followed by resin film infusion (RFI). The mechanical properties of the composites were studied,

and results show that the composites have significantly improved interfacial bonding and mechanical properties. © 2007 Wiley Periodicals, Inc. *J Appl Polym Sci* 104: 3588–3591, 2007

Key words: polytetrafluoroethylene; polymer-matrix composites; mechanical properties; interface; microstructure

INTRODUCTION

Continuous fiber reinforced polytetrafluoroethylene (PTFE) composites are a special and very important class of high performance materials with excellent dielectric properties, wide service temperature from -260 to 260°C , outstanding ablative resistance, and very high chemical resistance. This make them great potentially be used to produce high performance materials such as high frequency printed circuit boards (PCBs) or micro-wave dielectric materials, especially which are exposed to harsh environments including aeronautics and astronautics.^{1,2} However, the poor processing property limits its wide application. As we have known that PTFE has very high molten viscosity even at high temperatures and poor flowing property, so it is very difficult to use conventional techniques to fabricate continuous fiber reinforced PTFE composites. In fact, almost all PTFE based composites must be fabricated by sintering processing at high temperatures ($>300^{\circ}\text{C}$),^{3–5} and the processing procedure should be strictly abided by to guarantee the quality of the products, but it has been proved that it is difficult to completely avoid the formation of porosity resulting from the thermal pyrolysis of some organic materials,

therefore the resultant continuous fiber reinforced PTFE composites generally show weakened properties, especially the mechanical properties. In addition, because PTFE is highly chemical inert, so it is generally not possible to achieve strong interfacial bonding between PTFE matrix and fibers to ensure load translation. Above-mentioned problems are generally considered to be the main reasons of poor mechanical properties shown by continuous fiber reinforced PTFE composites.^{6,7}

How to address the issues has become a challenging assignment in the production of continuous fiber reinforced PTFE composites. In this study, a unique two-step method is reported for the first time to prepare densified continuous glass fiber reinforced PTFE composites with good interfacial adhesion and outstanding mechanical properties, the continuous glass fiber reinforced PTFE composite is coded as PTFE/GF composite. In detail, using conventional sintering to fabricate PTFE/GF composite, which is then used as the preformer and wet by a high performance resin via resin film infusion (RFI) technique because RFI technique has been identified as a cost-effective fabrication technique for producing damage tolerant textile composites.⁸

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EXPERIMENTAL

Materials

A high performance resin film (BDP) which is made up of *N, N'*-diphenylbismaleimide (BDM),

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o,o'-diallylbisphenol A (DBA), polyethersulfone (PES), and polyimide (PI) was selected as the RFI film due to its outstanding thermal properties, mechanical properties, hot-wet resistance, and dielectric properties which corresponds with outstanding properties of PTFE, it was prepared by our research group following the procedure reported in our previous literature.⁹ The typical properties of BDP were listed in Table I.⁹

PTFE aqueous dispersion with a solid content of 60 wt % (the molten point of the solid is 327°C) was bought from Shanghai Sanaifu, and was surface treated by γ -radiation following the procedure described in previous literatures.¹⁰ E-glass woven fabric (MW-100) was bought from Shaanxi Xingping Glass Fibers Factory (China).

Preparation of continuous glass fiber reinforced polytetrafluoroethylene composites

The densified continuous glass fiber reinforced PTFE composites were fabricated by following two steps: (1) E-glass woven fabric was impregnated by PTFE dispersion until the PTFE content was achieved, then the impregnated E-glass woven fabric was air-dried at 120–200°C for 1 h to form a prepreg. After that, the prepregs were cut into square plies and stacked in a mold to yield a laminate with the required dimension, and then the mold was put into a compressor for cool molding. The cool molding was processed with a pressure of 5–45 MPa according to the requirement for obtaining composites with different porosities, the dependence of porosity on the pressure is shown in Figure 1. It is worthy to note that it is very difficult to prepare PTFE/GF composites without porosity; our researches have shown that the PTFE/GF composite fabricated with the optimum processing factors (determined according to the mechanical properties) still contains 1.9% porosity. After that the compressed laminate was demolded. The resultant compressed laminate was sintered at 330°C/1 h + 380°C/2 h, then the resultant laminate was referred as the preformer of following RFI technique. (2) Sheet of BDP film was laid into the mold, and then the preformer obtained above was laid on the top of the film. The lay-up was vacuum bagged to remove air through the pre-

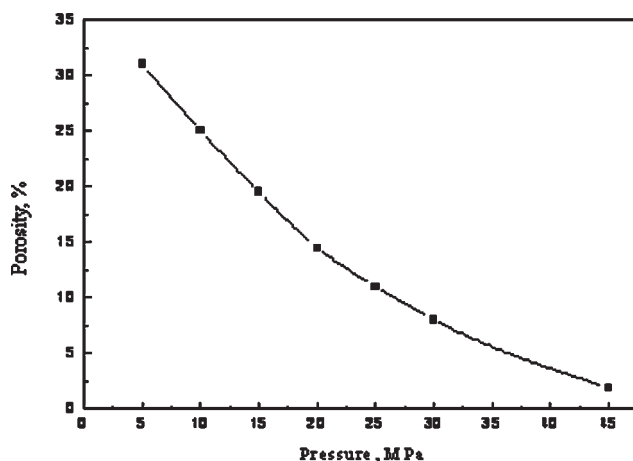


Figure 1 Dependence of porosity on the pressure for fabricating PTFE/GF performers.

former, and then was kept at 0.09 MPa at room temperature for 30 min; After that, the pressure was reduced to 0.026 MPa and the temperature was increased to 120°C. After maintaining that temperature for 2 h, the laminate was cured via a curing cycle of 150°C/3 h + 180°C/2 h + 200°C/2 h. Post-curing of the laminate was performed in an air-circulation oven at 220°C for 10 h.

Measurements

A Hitachi S-570 SEM was used to observe the tensile fracture morphology and interfacial adhesion of the samples. Instron 5567 universal tester was used to measure the flexural properties and interlaminar shear strength (ILSS) of composites according to ASTM D 790-00 and GB/T 1450.2-1983, respectively.

RESULTS AND DISCUSSION

It is known that the flexural property (especially the flexural strength) of a composite reflects the integrate properties of the composite because of its special stress loading and damage mechanic,¹¹ while ILSS of a composite is mainly depended on the strength of the matrix used and the interfacial adhesion between the matrix and reinforcements,¹² Therefore, flexural strength and ILSS have been selected as the typical properties to evaluate the integrate mechanical properties of a composite.

Figure 2 depicts the flexural strengths of PTFE/GF performers with various porosities and their corresponding BDP/PTFE/GF composites, the flexural strength of the PTFE/GF performer with the lowest porosity of 1.9% as well as the optimum mechanical properties is also included for comparison. It can be seen that (1) the flexural strength of each BDP/PTFE/GF composite is much higher than that of its

TABLE I
The Typical Properties of BDP Film

Tensile	
Strength (MPa)	90.2
Modulus (GPa)	4.3
Flexural	
Strength (MPa)	180
Modulus (GPa)	4.1
Impact strength, (kJ/m ²)	16.7

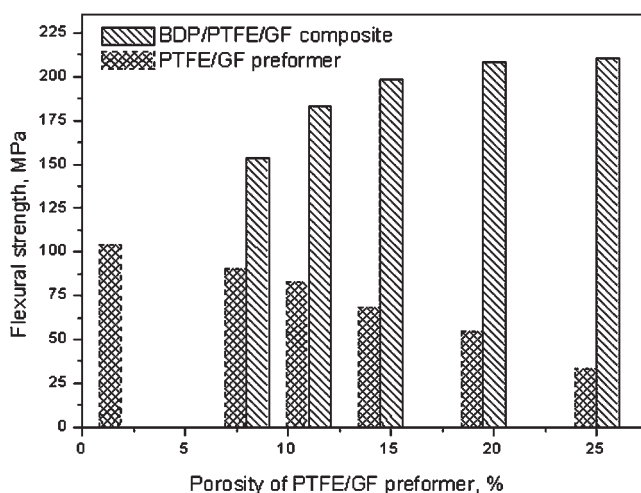


Figure 2 The flexural strengths of PTFE/GF preformers and their corresponding BDP/PTFE/GF composites.

PTFE/GF preformer, the former is about 1.7–3.8 multiple of the latter. In detail, if the PTFE/GF preformer has higher porosity, then its flexural strength is lower, and the flexural strength of its corresponding BDP/PTFE/GF composite is larger multiple of that of the preformer. (2) For BDP/PTFE/GF composites, the one based on the preformer with higher porosity has higher flexural strength than that based on the preformer with lower porosity, while in case of these composites based on the preformers with much higher porosity such as 20% or even higher, the values of their flexural strength are almost similar. The results could be interpreted that the BDP/PTFE/GF composite based on the preformer with higher porosity has more content of BDP resin, while BDP resin contains many reactive groups and thus has good bonding with glass fibers, so the addition of BDP resin tends to improve the interfacial adhesion between the matrix and fabrics, and the higher content of BDP resin in BDP/PTFE/GF composites, the more areas with good bonding along the interface are, and thereby leading to higher flexural strength.

The improvement of interfacial adhesion by BDP resin can be confirmed by comparing the SEM images of the interlaminar shear fracture surfaces of PTFE/GF composite and BDP/PTFE/GF composite as shown in Figure 3, it can be seen that there are many areas with nonbonding appear along the interface between PTFE matrix and glass fabric of PTFE/GF composite, contrast, above phenomena do not appear in BDP/PTFE/GF composite.

Figure 4 describes the ILSS values of BDP/PTFE/GF composites based on PTFE/GF preformers with various porosities, the flexural strength of the PTFE/GF preformer with the lowest porosity of 1.9% as well as the optimum mechanical properties is also

included for comparison. It can be seen that all BDP/PTFE/GF composites have higher ILSS values than the PTFE/GF preformer with the lowest porosity of 1.9% and the optimum mechanical properties, reflecting that BDP/PTFE/GF composites have better interfacial adhesion and improved concrete strength of the overall matrix than PTFE/GF composites do. In addition, the BDP/PTFE/GF composite based on the preformer with higher porosity has higher ILSS value than that based on the preformer with lower porosity, which also can be explained by the improvement of interfacial adhesion resulting from BDP resin as discussed earlier.

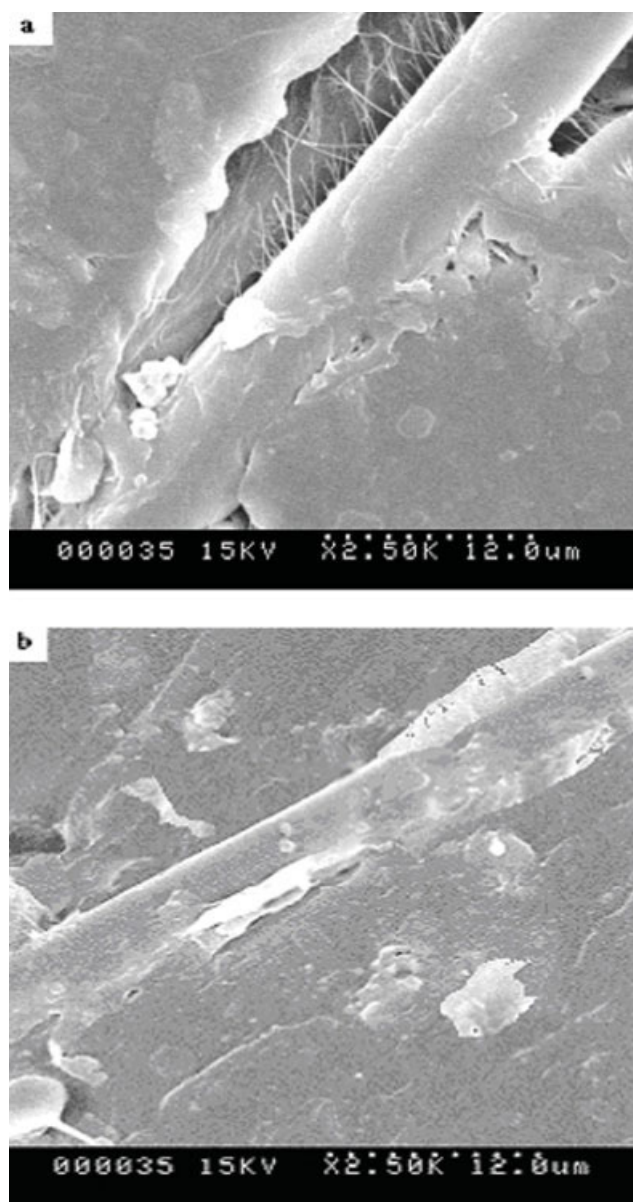


Figure 3 SEM images of the interlaminar shear fracture surface of PTFE/GF composite with a porosity 14.5 wt % (a) and corresponding BDP/PTFE/GF composite (b).

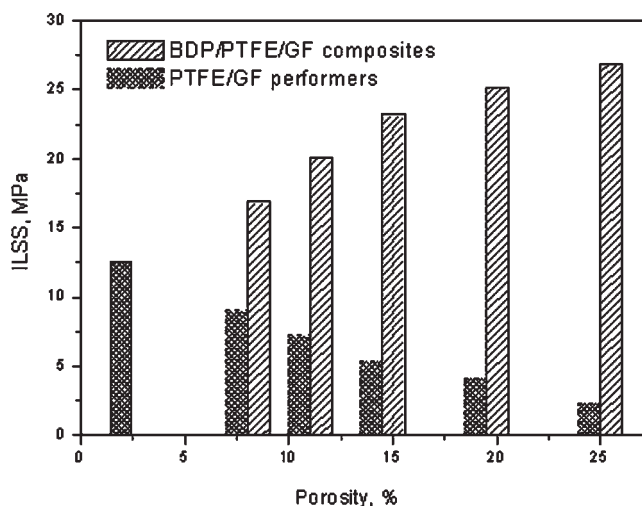


Figure 4 The ILSS of BDP/PTFE/GF composites based on PTFE/GF performers with various porosities.

CONCLUSIONS

In summary, an unique two-step technique for fabricating densified BDP/PTFE/GF composites with significantly improved interfacial bonding and mechanical properties has been developed. The BDP resin "fills" the porosity and defects of original PTFE/GF

composites prepared by traditional sintering processing, and improve the overall interface bonding between the matrix and fibers. The improved overall interface bonding leads to significantly improve the mechanical properties such as flexural and interlaminar shear strength of fiber reinforced PTFE composites.

References

- Guang, C. S.; Wang, W. S.; Fluoropolymers and Their Applications; Chemical Engineering Press: Beijing, China, 2004; Chapter 2.
- Aglan, H.; Gan, Y.; El-Hadiq, M.; Faughnan, P.; Bryan, C. J. *Mater Sci* 1999, 34, 83.
- Jog, J. P. *Adv Polym Technol* 1993, 12, 281.
- Klaas, N. V.; Marcus, K.; Kellock, C. *Tribolo Int* 2005, 38, 824.
- Andena, L.; Rink, M.; Polastri, F. *Polym Eng Sci* 2004, 44, 1368.
- Joyce, P. J.; Joyce, J. A. *Int J Fract* 2004, 127, 361.
- Fu, G. D.; Zhang, Y.; Kang, E. T. *Adv Mater* 2004, 16, 839.
- Han, N. L.; Suh, S. S.; Yang, J. M.; Hahn, H. T. *Compos A* 2003, 34, 227.
- Liang, G. Z.; Wang, D. *J Appl Polym Sci* 2001, 81, 2918.
- Lunkwitz, K.; Burger, W. *J Appl Polym Sci* 1996, 60, 2017.
- Song, H. C. *Polymer Composites*; Beijing University of Aeronautics and Astronautics Press: Beijing, China, 1985; Chapter 5, p 156.
- Liang, G. Z.; Hu, X. L. *Polym Int* 2004, 53, 670.