MANUFACTURING CARBON NANOTUBES TOUGHENED GLASS FIBER/EPOXY RESIN LAMINATES USING DOUBLE VACUUM ASSISTED RESIN INFUSION MOLDING

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Keywords: *Fiber reinforced composite laminate, MWCNTs, Epoxy resin, VARIM, Mechanical property*

Abstract

In this paper, a kind of multi-walled carbon nanotubes (MWCNTs) with 0.05wt% was added in an epoxy resin, and unidirectional glass fiber fabric/epoxy resin laminates were prepared using vacuum assisted resin infusion molding (VARIM). In order to promote the toughness effect of the MWCNTs on the composites, a modified VARIM, named double vacuum assisted resin infusion molding (DVARIM) was devised to optimize the process procedures, which can improve the resin infiltration especially in the fiber bundles. The defects of the laminates fabricated with the two processing methods were examined, and the mechanical properties including flexural property and interlaminar shear strength (ILSS) were measured. Furthermore, the enhancement mechanism of MWCNTs for the laminates was analyzed. It shows that the mechanical properties of the laminates with MWCNTs processed by DVARIM are obviously higher than those with and without MWCNTs processed by VARIM. Moreover, the infusion process influences the fiber/resin interfacial bonding, which is attributed to the difference of the distribution of CNTs in the laminates.

1 Introduction

Due to high strength, modulus, surface area and aspect ratio, carbon nanotubes (CNTs) are excellent candidates for nano-reinforcing polymers and polymer matrix composites. Recently, CNTs reinforced fiber/thermoset resin composites have attracted great interests, because they can simultaneously take advantages of the properties of micro-material (such as glass fiber and carbon fiber) and nano-material. The reported studies use different processing techniques for manufacturing CNTs/fiber/resin composites, including autoclave, RTM, and VARIM processes [1-4]. Vacuum assisted resin infusion molding (VARIM or named VARTM) is an important low-cost manufacturing method, which injects resin into fiber fabrics using vacuum with only one-side mold. With the assistances of high permeable media and pipelines, VARIM can produce high-performance composites with low porosity and high producing efficiency. To date, VARIM has been widely used to manufacture large size fiber reinforced composite parts, such as wind turbine blade, ship hull and aircraft structure. To further enhance the mechanical performances of the produced parts processed by VARIM, adding CNTs in resin has been considered as a promising means [3-7].

Zhou et al [5] studied the effect of adding CNTs on the mechanical property of carbon fabric/epoxy composites in VARIM, and the improvements in flexural strength, glass transition temperature, and decomposing temperature were observed. Fan et al [6,7] fabricated glass/epoxy composites with oxidized multi-walled carbon nanotubes dispersed in the epoxy. The introduction of CNTs into the composites increased the interlaminar shear strength by up to 33%. Moreover, in order to increase the concentration of CNTs in the resin, they adopted double vacuum assisted resin transfer molding to reduce the compaction of the fiber bed and ensure high permeability during injection. Chandrasekaran [4] and Fan [6] indicated that the process designs of VARIM had strong impacts on the dispersion and orientation of CNTs in composites, which determined the enhancing degree of CNTs.

In this paper, to study the effect of process design of VARIM, a kind of multi-walled CNTs (MWCNTs) with 0.05wt% was added in an epoxy resin, and unidirectional glass fiber fabric/epoxy resin laminates were prepared by VARIM and double vacuum assisted resin infusion molding (DVARIM). The mechanical properties including flexural property and interlaminar shear strength (ILSS) of composite laminates with the two processing methods were examined, and the enhancement mechanism of CNTs for the laminates was analyzed.

2 Experimental

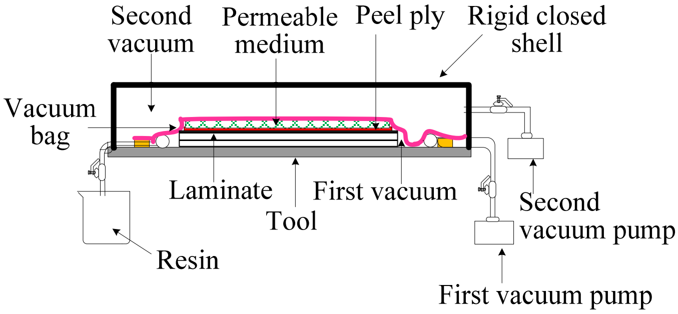
2.1 Materials

The resin used in this study was a commercially available HJ-966 epoxy resin system supplied by Dasen Inc. It is a low-viscosity epoxy resin with modified amine curing agent. A kind of hydroxylating MWCNTs supplied by Bayer Inc. was adopted, which diameters were about 13 nm and the length was more than 1 μm. The unidirectional glass fiber fabrics were produced by Jushi Inc. and had a fiber density of 1200 g/m^2.

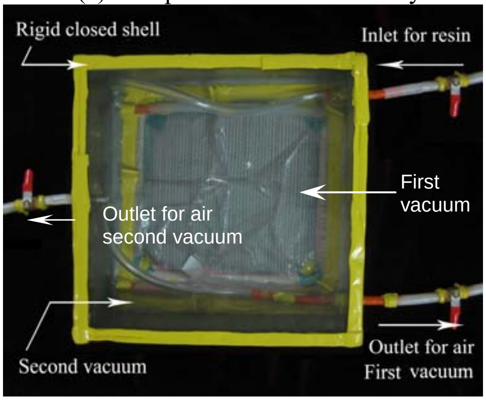
2.2 Manufacturing

The MWCNTs/epoxy suspension was prepared using ultrasonic agitation for 6h, and the content of MWCNTs in pure resin was 0.05wt%. The resin used for the raw sample without the MWCNTs, was also subjected to identical sonication process. Four layers of 20 cm×20 cm glass fiber fabrics were cut and sealed under a vacuum bag. As shown in Fig. 1(a), a layer of high permeable medium was put on the upper fabric, and a peel ply between the permeable medium and the fabric was to facilitate separating them after cure. The air was evacuated by applying a vacuum to the outlet pipe, and the resin in a pot was injected into the fiber preform through the high permeable medium at ambient temperature.

For the VARIM, only the first vacuum pump was used, and the fiber preform compacted under the external atmospheric pressure acting on the bag. For the DVARIM, the resin was injected under the drive of the first vacuum pump, and after 5 min the first vacuum line was clamped and the injecting pipe was shut. Then, the outlet of the second vacuum line was opened, and the entire rigid closed shell was under slightly higher vacuum than the first vacuum (vacuum in the vacuum bag) for 5 min. This slightly higher second vacuum released the external atmospheric pressure induced by the first vacuum, and allowed the compressed preform to relax and recover some of its original thickness. This recovery opened up the gaps among the fiber bundles and among the fiber filaments, making it easier for the MWCNTs/epoxy suspension to impregnate and flow into the preform. Finally, the second vacuum was released and the first vacuum line was reopened to reapply the external atmospheric pressure onto the preform for achieving the desired fiber content about 73wt%. Fig. 1 shows the assembly of the two process designs. All composite laminates were cured in oven at 70°C for 6 h.



(a) Components of the assembly



(b) Photograph of the assembly

Fig. 1 Schematic of double vacuum assisted resin infusion molding.

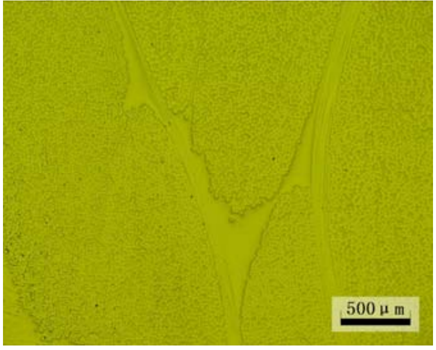
2.3 Characterization

In this study, we used the flexural property and ILSS to evaluate the effects of the addition of MWCNTs on the composites. The flexural modulus and strength were measured according to GB/T 1449-2005, and ILSS test was conducted based on GB/T 1450.1-2005. Defects in the laminates were studied from micrographs, and the fracture cross-sections of the laminates after mechanical testing were analyzed under SEM.

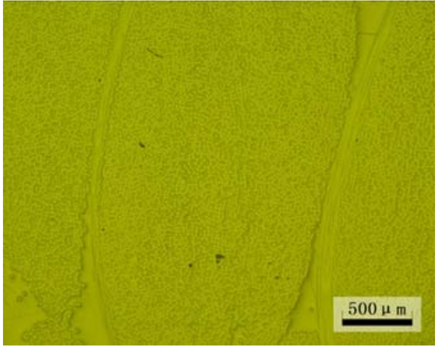
3 Results and discussion

The unidirectional glass fiber fabric/epoxy resin laminates with MWCNTs were prepared using the VARIM and DVARIM processes. For studying the effect of MWCNTs, the laminates without MWCNTs were also prepared using the two processes. According to the results from ablation method, all prepared laminates have the almost same fiber contents, which range from 73wt% to 74wt%.

Fig. 2 shows the typical micrographs on the cross-sections of the laminates with CNTs manufactured with the VARIM and DVARIM processes. It can be seen that there are no voids and other defects in the laminates for both the processes. The distributions of fiber bundles and fiber filaments in the bundles are similar for different laminates. The resin at 25 °C has low viscosity (200mPa·s) and low surface tension (25mN/m), and does not change obviously after adding 0.5wt% MWCNTs. Thus, the effect of MWCNTs on the resin infiltration into the fiber fabrics is supposed to be negligible.



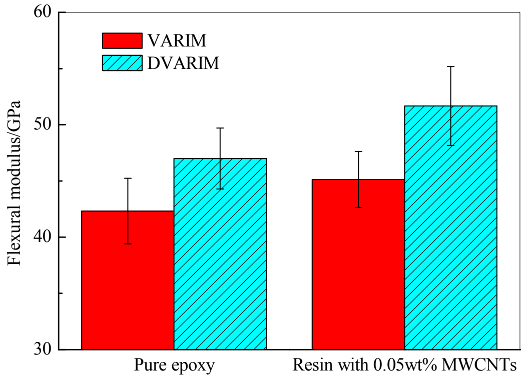
(a) VARIM



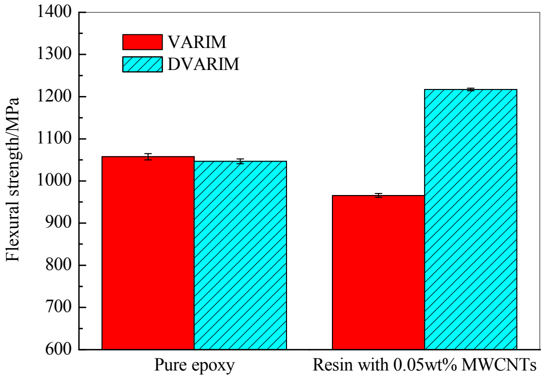
(b)DVARIM

Fig. 2 Micrographs of glass fiber/epoxy resin composite laminates with MWCNTs manufactured by different resin infusion methods.

The flexural modulus and strength of the laminates with MWCNTs processed with VARIM and DVARIM are given in Fig.3. For the purpose of comparison, the flexural properties of the laminates without MWCNTs are also shown in Fig.3. The laminates processed with DVARIM have higher flexural modulus, especially for the samples with MWCNTs. An approximately 22.1% increase in flexural modulus is obtained for the samples with MWCNTs processed by DVARIM, compared to the samples without MWCNTs processed by VARIM, while the increase is about 11.1% for the samples without MWCNTs processed by DVARIM. Moreover, adding MWCNTs does not offer the enhancement in flexural strength for VARIM, but the flexural strength is increased by 15.1% for DVARIM. The similar results are also obtained for ILSS test. Fig.4 demonstrates the ILSS of the laminates with the two processes. There is no obvious difference in ILSS for the samples with and without MWCNTs under VARIM. However, the MWCNTs enhance the samples using DVARIM with a 10.9% ILSS improvement.



(a) Flexural modulus



(b) Flexural strength

Fig. 3 Flexural properties of glass fiber/epoxy resin composite laminates with and without MWCNTs manufactured by different resin infusion methods.

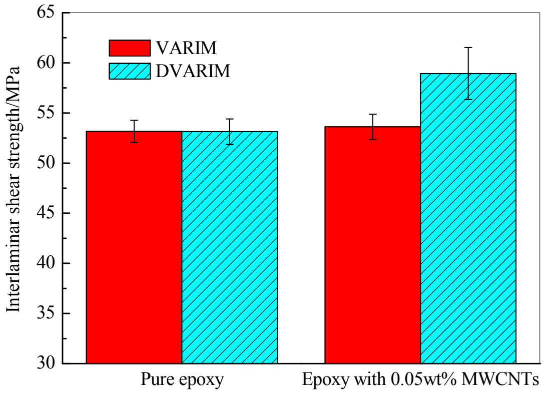
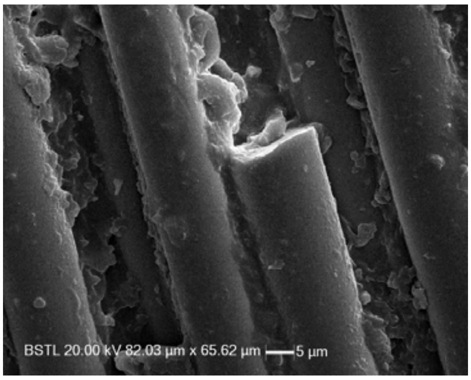


Fig. 4 Interlaminar shear strength of glass fiber/epoxy resin composite laminates with and without MWCNTs manufactured by different resin infusion methods.

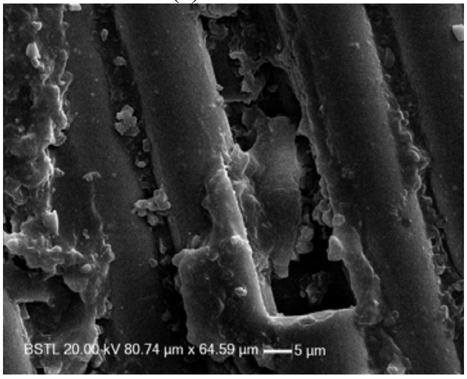
In our studies, the content of MWCNTs is very low, which is 0.05wt% in the pure resin and is 0.01wt%

in the composite laminates. Therefore, the enhancement of the MWCNTs on the resin matrix is limited, and the improvement of the MWCNTs on the mechanical properties of the composites using DVARIM is attributed to the effect of MWCNTs on the interphase between resin and glass fiber.

Because the spaces among the fiber bundles and among the filaments are larger during DVARIM, the filtering effect on MWCNTs caused by the compaction of the preform under the application of the vacuum is not prominent [6]. It makes it easier for the MWCNTs to disperse on the surfaces of the fibers and form bridges between matrix and fibers. This process would increase the interfacial bonding strength, so the flexural property and ILSS increase significantly, as shown in Fig. 3 and Fig. 4. The SEM of fracture cross-sections of the laminates after mechanical testing verifies the hypothesis, as given in Fig. 5. It can be seen that there is lots of resin left on the fiber surfaces for the samples with MWCNTs under DVARIM, and the fiber surfaces are smooth for the samples with MWCNTs under VARIM, demonstrating stronger interfacial bonding for DVARIM.



(a) VARIM



(b)DVARIM

Fig. 5 SEM of fracture cross-sections of glass fiber/epoxy resin composite laminates with MWCNTs manufactured by different resin infusion methods.

4 Conclusions

This study was conducted to determine whether the process designs of VARIM have great impacts on the improvement on the mechanical property of glass fiber reinforced composites by adding low content MWCNTs in epoxy resin. For common VARIM, the addition of MWCNTs only increases the flexural modulus, and has negative or no influence on the flexural strength and ILSS. However, these mechanical properties of the laminates with MWCNTs are obviously improved for DVARIM. According to the SEM of fracture cross-sections of the laminates, stronger interfacial bonding for DVARIM is found, compared to the case for VARIM. Therefore, we conclude that the infusion process influences the fiber/resin interfacial bonding, which is attributed to the difference of the distribution of CNTs on the fiber surfaces during the molding process.

5 Acknowledgments

This work was supported by funding from the National 863 Program of China (Grant No. 2009AA0345).

References

[1] E. Garcia, B. Wardle and A. Hart “Joining prepreg composite interfaces with aligned carbon nanotubes”. *Compos Part A*, Vol. 39, No. 6, pp 1065–70, 2008.

[2] M. Wichmann, J. Sumfleth, F. Gojny, M. Quaresimin, B. Fiedler and K. Schulte “Glass–fibre-reinforced composites with enhanced mechanical and electrical properties – benefits and limitations of a nanoparticle modified matrix”. *Eng Fract Mech*, Vol. 73, No. 16, pp 2346–59, 2006.

[3] L. Boeger, M. Wichmann, L. Meyer and K. Schulte “Load and health monitoring in glass fibre reinforced composites with an electrically conductive nanocomposite epoxy matrix”. *Compos Sci Technol*, Vol. 68, No. 7-8, pp 1886–94, 2008.

[4] V. Chandrasekaran, S. Advani and M. Santare “Role of processing on interlaminar shear strength enhancement of epoxy/glass fiber/multi-walled carbon nanotube hybrid composites”. *Carbon*, Vol. 48, No. 13, pp 3692-9, 2010.

[5] Y. Zhou, F. Pervin, L. Lewis and S. Jeelani. “Fabrication and characterization of carbon/epoxy composites mixed with multi-walled carbon nanotubes”. *Materials Science and Engineering A*, Vol. 475, No. 1-2, pp 157–65, 2008.

[6] Z. Fan, M. Santare and S. Advani “Interlaminar shear strength of glass fiber reinforced epoxy composites enhanced with multi-walled carbon nanotubes”. *Composites: Part A*, Vol. 39, No. 3, pp 540–54, 2008.

[7] Z. Fan, K. Hsiao and S. Advani “Experimental investigation of dispersion during flow of multi-walled carbon nanotube/polymer suspension in

fibrous porous media". *Carbon*, Vol. 42, No. 4, pp 871-6, 2004.