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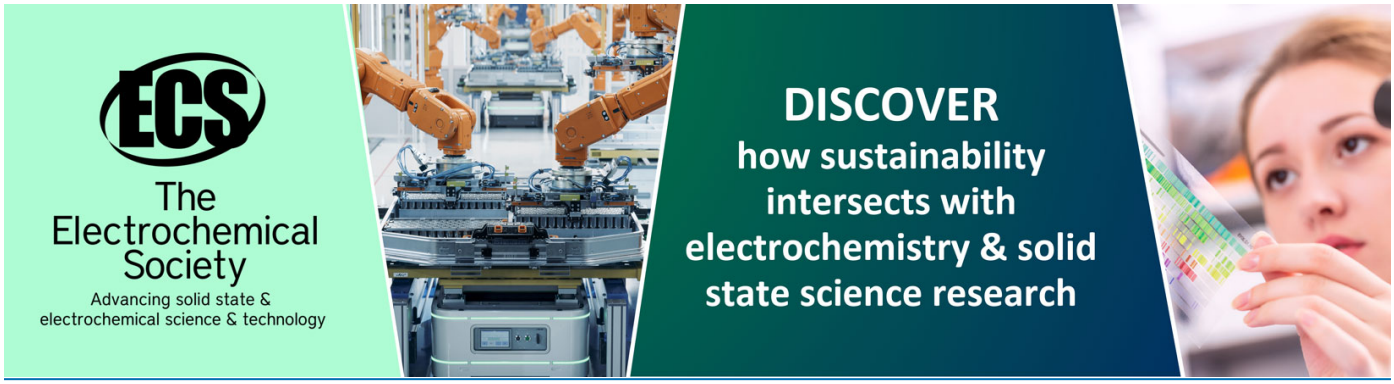

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Effect of fibre sizing on the interlaminar properties of polyamide matrix composites

G Szebényi[1] and B Magyar[2]

[1]Assistant professor, BME, Faculty of Mechanical Engineering, Department of Polymer Engineering, Budapest, Hungary
[2]PhD student, BME, Faculty of Mechanical Engineering, Department of Polymer Engineering, Budapest, Hungary

E-mail: szebenyi@pt.bme.hu

Abstract. The aim of our study was to compare the interlaminar performance of PA based composites with different fiber sizing to investigate the importance of proper sizing. Composites were prepared by film-stacking from reinforcing materials with three available sizings compatible for vinylester, epoxy and polyamide. The composites were compared through standard static and self-developed dynamic and cyclic interlaminar shear test methods and scanning electron microscopy. The results showed that the proper sizing is crucial for the good mechanical performance of the composites, the PA sized fiber composites significantly better in all mechanical properties.

1. Introduction

Nowadays the application of fiber reinforced composites is spreading in the fields of both high performance materials [1, 2] and commercial applications [3, 4]. While in the previous decades long fiber reinforced composites were only applied in demanding low volume applications or mainly as covering materials for large volume production [5, 6], the tendency is shifting towards the use of long fiber reinforced composites in demanding large volume applications as load bearing structures [7, 8] and automotive structural parts [9, 10]. While the relatively high curing time of thermoset matrices was an obstacle, the emerging of high output technologies like HP-RTM (high pressure resin transfer moulding) [11] or T-RTM (thermoplastic resin transfer moulding) [12] can provide a low cycle time of some minutes compared to hours enabling the use of these materials in areas where thousands of parts have to be annually produced [13]. While these technologies open new ways for composites, some problems of these materials also become more emphasized. In standard thermoset materials proper sizing of the fibers gives adequate adhesion between the fibers and the matrix. Achieving such a level of adhesion [14, 15] is more challenging when using thermoplastic matrix materials, where in some cases chemical reactions are completely absent during production. In the application areas of most of these composites, the presence of constant vibration and dynamic loads can endanger the structural integrity of the composites by the formation of fiber-matrix debonding and delamination in the loaded areas. In all of the applications the interlaminar properties of the composites is a key factor for static and dynamic load bearing capability, failure method and fatigue cycle life. While the most reinforcing materials come with surface sizing developed for different kinds of thermoset matrices, where these are mostly used, some specific grades are already marketed with sizing especially selected for PA matrix applications.

The aim of our study was to compare the interlaminar performance of PA based composites with different fiber sizing to investigate the importance of proper sizing. The composites were compared through standard static and self-developed dynamic and cyclic interlaminar shear test methods and scanning electron microscopy.

2. Materials and Methods

2.1. Materials, composite production

The matrix material applied for the film-stacking method for the composite production was Schulamid 6 MV13 NATUR polyamide 6, similar to the caprolactam-based in situ systems used for the T-RTM production technology. The PA films were extruded by a Labtech Scientific film extruder line. The extruder zone temperatures were 230, 235, 245 and 250°C from the hopper to the die respectively. The screw rotational speed was 30 rpm. The three zone temperatures of the film die were set to 260°C. The pickup rolls in the production line were set to provide 1.1 m/minute and 1.7 m/minute pull speed at the rubber pickup rolls. The winding roll was set to 9 rpm. 155 mm wide and 150±10 μm thick film was extruded. The thickness of the film was measured by a Mitutoyo Digimatic digital micrometer.

Three reinforcement materials were selected for the specimen production with glass fibers with different types of fibre sizing prepared to be used for vinylester (VE), epoxy (EP) and polyamide (PA) by Saertex (Germany). All reinforcement materials had the same plain woven structure with a surface weight of 300 g/m^2 composed of 1200 tex glass rovings.

The composite sheets were produced by film-stacking method using a Schwabenthan Polystat 300S platen press. The platen temperature was set to 260°C on both sides according to the results of the preliminary tests for optimal impregnation. The produced composite layup consisted of 6 layers of reinforcement material at uniform 0/90 direction and 7 layers of matrix. The pressing was performed at 300 bars piston pressure, which resulted in 53.7 bar pressing pressure on the 310x310 mm composite plate surface. The cooling was started after the completion of the 3 minutes pressing cycle. The pressing was sustained during the whole process. The platens were opened at 60°C temperature.

Interlaminar shear strength (ILSS) test specimens were cut out of the laminates in the 0° direction according to ASTM D 3846 – 94 by a Mutronic Diadisc 4200 disc saw. The notches on the opposite sides of the specimens were also machined by the same device. The specimen geometry was defined according to ASTM D 3846 – 94, with notches on the opposite sides of the specimens 6.4 mm from eachother. The notch thickness was 1 mm (as a result of the diamond disc thickness), the notch depth was half of the laminate thickness to achieve a stable delamination in the laminate center plane.

2.2. Test methods

The interlaminar properties of the composites were investigated by a static, a dynamic and a cyclic method.

The static method used for the interlaminar characterization was the double notched (DNS) interlaminar shear test method according to ASTM D 3846 – 94 presented in Figure 1. The test was performed using a Zwick Z020 universal computer controlled tensile tester at 1.3 mm/minute crosshead speed.

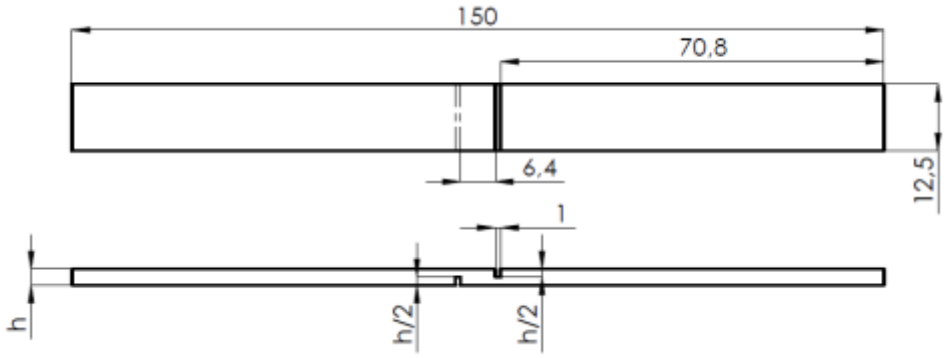

Figure 1. Double notched specimen

The dynamic method was a self-developed dynamic interlaminar shear test method [16] based on the static standard. The standard specimen was tested using a Ceast Resil Impactor Junior pendulum equipped with a 15 J tensile impact hammer. The specimen was fixed in the anvil of the tester and loaded through the 30 g weight crosshead attached to the other end of the specimen. The starting angle was 150° resulting in an impact velocity of 3.7 m/s. The energy required for the fracture was recorded by the machine. The energy required for the movement of the crosshead was deducted from the energy value. The resulting dynamic interlaminar shear strength was calculated from the corrected fracture energy divided by the shear cross-section between the notches.

The cyclic interlaminar shear tests were also carried out according to a self-developed method [16]. The standard specimens were investigated using an Instron 8872 computer controlled servo-hydraulic fatigue tester. The test was performed in displacement-controlled operation. The applied loading waveform was pulsating sinusoid with a frequency of 5 Hz with peak values set to the displacements corresponding to 50 and 10% of the failure displacements recorded during the static tests.

The scanning electron microscope (SEM) micrographs were taken using a JEOL JSM 6380LA microscope in high vacuum operation. The surfaces of the samples were gold spur coated prior to the investigations by a JEOL FC 1200 device to achieve the electrical conductivity of the surface to avoid local charge accumulation.

3. Results and Discussion

3.1. Static interlaminar properties

The results of the static interlaminar shear tests are presented in Figure 2. From the results the requirement of proper sizing can be clearly seen. The samples with VE and EP sizing showed poor interlaminar shear strength compared to the composite with proper PA sizing. The EP sizing slightly outperformed the VE sizing. Comparing the interlaminar shear strength values to the literature data [17, 18] it can be seen that even in case of the PA sized reinforcement, the interlaminar shear strength is inferior compared to the thermoset system results. This can be caused by the lack of chemical reactions during processing: no covalent bonds can be formed between the fibers and the matrix. Secondly even in case of thermoplastic matrices with relatively low melt viscosity the low shear speed of film-stacking can not force the melt into the reinforcing fiber rowings for proper impregnation. The T-RTM process, creating PA in situ from caprolactam can address both these problems, the monomer viscosity is less than the viscosity of most thermoset resins and because of the in situ process the matrix can covalently bond to the reinforcement sizing.

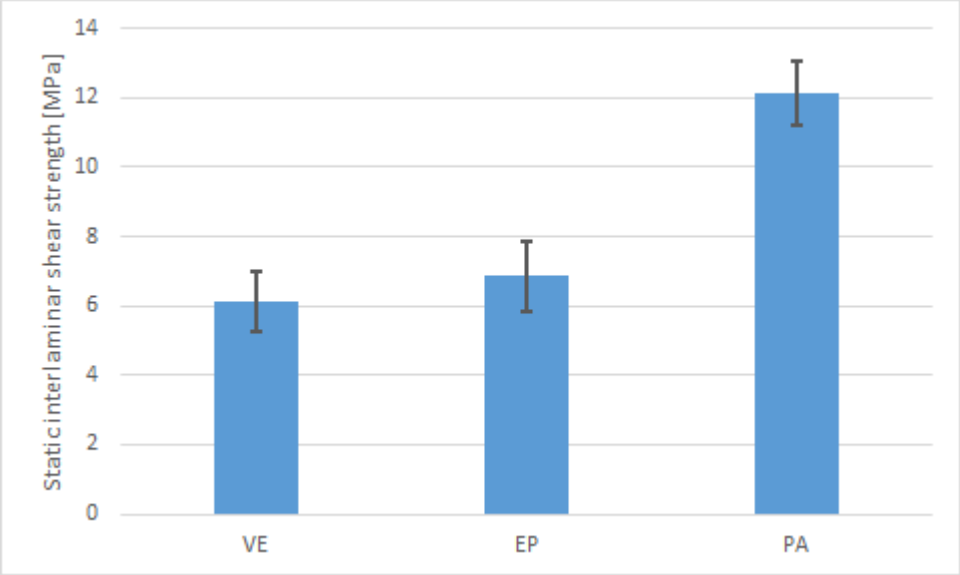

Figure 2. Results of the static interlaminar shear tests
(VE- fiber sizing for VE, EP – fiber sizing for EP, PA – fiber sizing for PA)

The results of the dynamic interlaminar shear tests are presented in Figure 3. Here, the impact produces a much higher strain ratio, the crack has to propagate much faster, the ductile matrix behaves much more brittle than in case of static loading. The targeted automotive sector requires high ductility of most of the composite parts, ductile behavior means more energy absorption in case of an accident, and less sharp fracture edges, which can cause injuries to passengers and pedestrians. In the results the clear superiority of the PA sizing is dominant. When comparing to standard Charpy strength results measured at similar neat PA materials [19], the dynamic interlaminar shear strength is much lower, especially with improper sizing. By constructing the T-RTM produced parts with neat PA edges, their performance can be probably greatly improved: the edges show much more ductile behavior and they can also hinder the crack to initiate at the free edges of the reinforced composite.

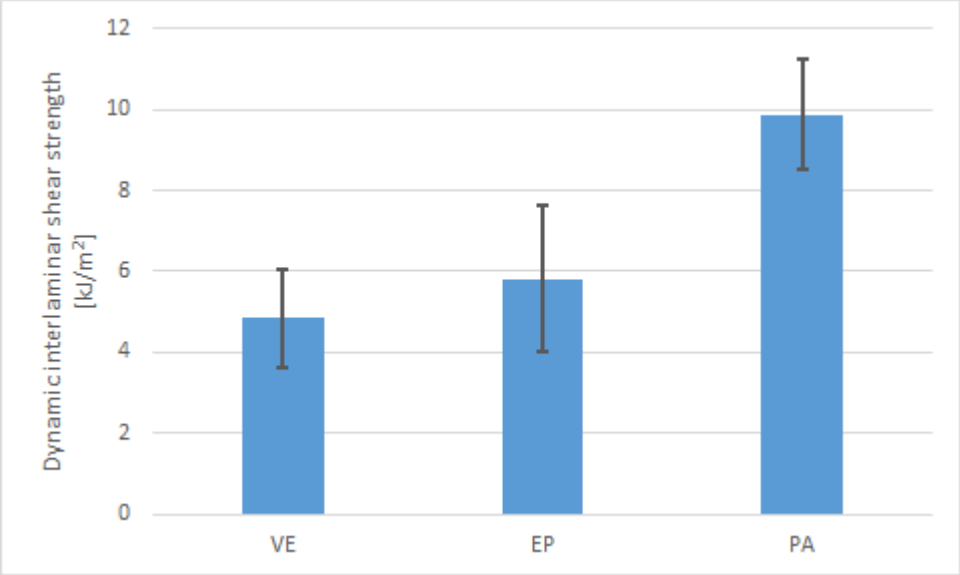

Figure 3. Results of the dynamic interlaminar shear tests
(VE- fiber sizing for VE, EP – fiber sizing for EP, PA – fiber sizing for PA)

The results of the cyclic interlaminar shear tests are presented in Figure 4. The PA sized fabrics performed much better. The load levels were defined based on the static interlaminar shear strength of the individual materials, so in case of the PA sized system the applied load was approximately two times higher compared to the other systems. Considering this, the performance of the PA sized system is even better. In case of fatigue loading of composites, the main failure process is not the propagation of an individual crack, but the formation of numerous micro-cracks in the system at the weakest points, which are most of the cases at the fiber-matrix interface. By improving the adhesion with proper sizing, the formation of these cracks can be avoided or at least postponed.

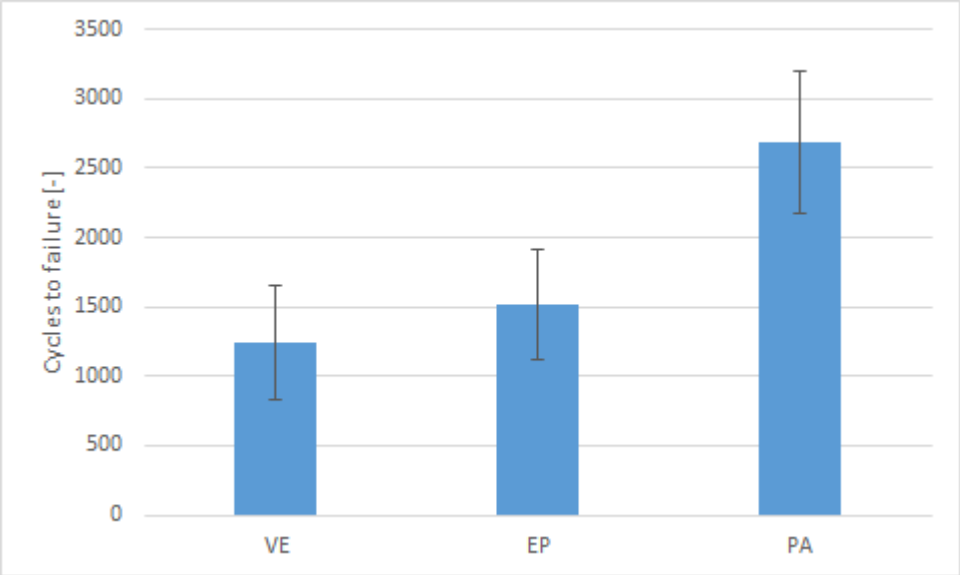

Figure 4. Results of the cyclic interlaminar shear tests
(VE- fiber sizing for VE, EP – fiber sizing for EP, PA – fiber sizing for PA)

The SEM micrographs of the different composites are presented in Figure 5. There is no significant difference in the structure of the composites. In all cases some trapped-in air voids and poorly impregnated rowings can be observed, which are common in composites produced by film-stacking. Although the mechanical performance showed clear difference in the adhesion between the fibers and the matrix, poor wetting or fiber matrix debonding at the interface are only present occasionally at all of the composites.

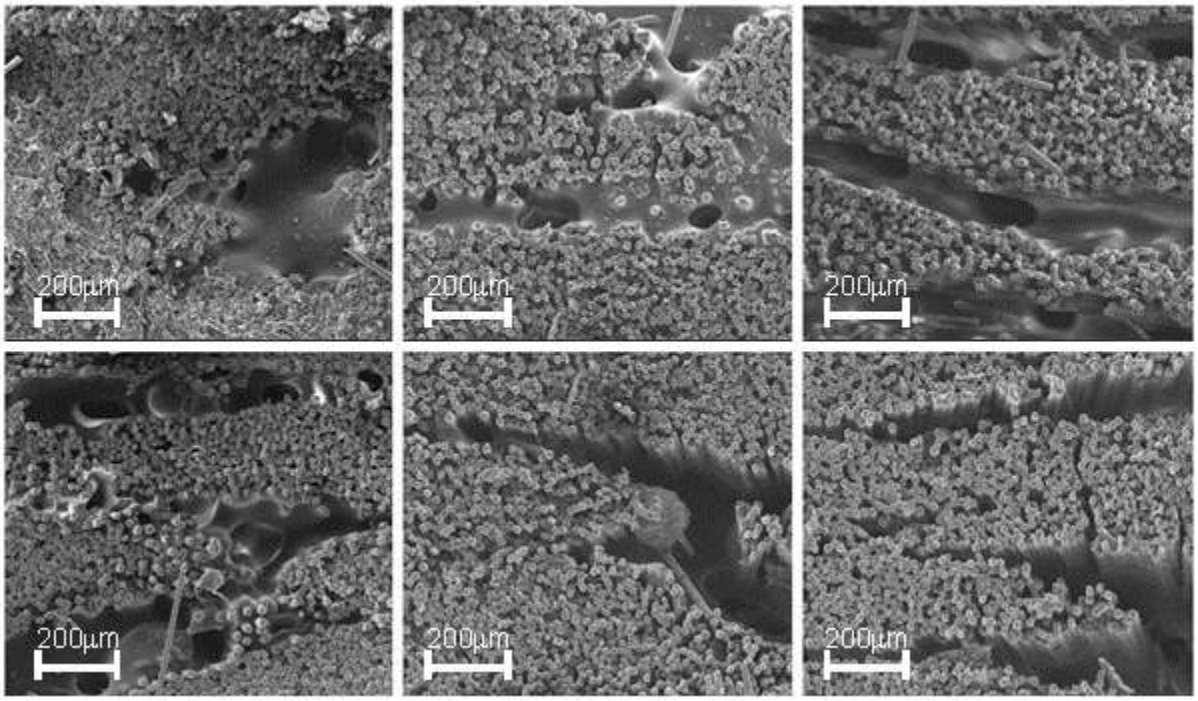

Figure 5. SEM micrographs of the composites containing reinforcement fibers with VE (left), EP (center) and PA (right) compatible sizing

4. Conclusions

According to the test results the proper fiber sizing is of key importance for the tested composite systems. In case of all, static, dynamic and cyclic interlaminar properties, the proper sizing showed significantly better mechanical performance and lifetime. Although the polar chemical structure of the binding groups of PA is similar to the structure of common thermoset resins used for reactive technologies in the composite industry, but the molecular chains, which are non-polar are much longer between the binding groups so the intermolecular forces will be significantly lower. Also the impregnation capability of the thermoplastic melt are much lower than in case of a thermoset resin, so more dry fiber bundles and trapped-in air voids have to be considered. With the rapid growth of the application of the reactive T-RTM technology more and more reinforcement manufacturers enter the market of PA compatible sized reinforcement materials, each with their own sizing composition. The methodology presented in our paper is capable for the determination of both static and cyclic interlaminar properties and the evaluation of new fiber sizing types.

Acknowledgements

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