Materials Science and Engineering A363 (2003) 335–344

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Compressive mechanical behaviour of E-glass/polyester composite laminates tailored with a thermoplastic preforming binder

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Received 2 May 2003; received in revised form 30 July 2003

Abstract

Compressive mechanical behaviour and failure modes of E (electrical)-glass/polyester composite laminates tailored with a thermoplastic preforming polyester binder were investigated under ply-lay up and in-plane loading directions. Fiber preforms with various amount of the binder were consolidated under heat and pressure. The preform compaction experiments were performed by applying compressive pressure to the preforms, and the average thickness as a function of pressure was measured. It was found that the highest compaction of the preforms and therefore the highest fiber volume fraction can be obtained with 3 wt.% of the binder. Further increase of the amount of binder decreases the degree of compaction. Composite panels were fabricated by vacuum-assisted resin transfer molding using fabric preforms with various binder concentrations. The present investigation reveals that there are considerable effects of the binder on the compressive mechanical behaviour of the composites. Compression testing of the composites showed that the average strength values are in the range of 400–600 and 150–300 MPa for ply-lay up and in-plane directions, respectively. Also, both the strength and modulus values increase up to 3 wt.% of the binder, and these values decrease with further addition of the binder. Scanning electron microscopy showed that failure modes of the composites are altered significantly by the presence of the binder. Furthermore, the interaction between the binder and the reacting resin was followed to determine the extent of the binder dissolution and its effects on the viscosity of the resin and the mechanical behaviour of the matrix polymer. The results indicate that there is a partial dissolution of the binder within the matrix resin.

Keywords: Polymer matrix composites; Compressive mechanical testing; Preform; Fracture

1. Introduction

Liquid molding (LM) processes such as resin transfer molding (RTM) and vacuum-assisted resin transfer molding (VARTM) have received considerable attention to manufacture high performance composite parts especially for automotive, civil engineering and military applications. RTM and VARTM processes have some unique advantages over all other composites manufacturing techniques [1–4]. These techniques offer an opportunity to produce large and complex shapes with the desired fiber orientation into a single molding via fiber preforms that may have the shape and dimensions of the final parts [2–4]. Fiber preforms can be made either by introduction of the binder over the glass fabrics to bind them together or by weaving, braiding, knitting or stitching continous fibres [1–3]. The recent studies showed that net shape thermoformable preforms can be produced by introduction of plastic binders between reinforcement fabrics to supply brief compaction [1–5]. In the fabrication of preforms, the plastic binder is uniformly spread over the surface of glass mats and then melted to stick on the surface. In the next stage, the desired number of binder-coated plies can be stacked together under application of heat and pressure. It was shown that final composite properties and microstructure depend to a great extent on compaction of the fiber preform to the designed thickness [8–11]. Furthermore, the degree of the compaction of the fiber preform has some significant effect on the permeability of the reinforcement during resin infusion and mutually fiber volume fraction and porosity. Therefore, an understanding of behaviour of the textile preforms under compressive forces is also of particular interest to be considered in the

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doi:10.1016/j.msea.2003.08.005

manufacturing of high performance composite parts [6–12]. It was shown that the distribution of plastic binders over the glass fabric surface affect the degree of compressibility of fiber preforms [6]. In addition, binder dissolution in the matrix resin may have some effects on viscosity profile of the matrix resin during mold filling. Furthermore, it was investigated in several studies that the introduction of the binder into the interlaminar region has some significant effects on the physical, mechanical and impact properties of the composites [1,5,14,15]. In the previous work [5], the effects of the preforming binder on the mechanical and ballistic performance of E (electrical)-glass/polyester composite systems were investigated. It was found that the peel strength of the preforms is increased with increasing binder content. The results also revealed that the flexural strength and modulus, Mode I interlaminar fracture toughness, ballistic performance and failure damage due to ballistic impact of the composite laminates are considerably affected by the presence of the binder. To our knowledge, there is no report in the literature showing the effects of the powdered preforming binders on the compressive mechanical properties of the composites loaded in ply-lay up and in-plane directions. It is known that the compressive strength of most composite laminates is much less than their tensile strength [13]. The evaluation of the compressive mechanical behaviour of the composites is critical since the compressive strength is one of the limiting factors in the design of composite structure.

The present study aims to investigate the compressive mechanical behaviour of the E-glass fiber/polyester composites tailored with a thermoplastic preforming binder. The glass fiber preforms were consolidated under heat and pressure using various binder concentrations. The effect of the binder on the preform compaction behaviour of the fabrics with the applied compressive force was also evaluated. The composite panels were fabricated by VARTM technique. Compression tests along the ply-lay up and in-plane directions were performed on the specimens sectioned from the panels with and without binder to determine the effects of preforming binder on the compressive behaviour of the composites. Furthermore, interactions between the thermoplastic binder and the thermosetting polyester matrix were determined to follow the extent of the binder dissolution in the reacting resin.

2. Experimental

2.1. Materials

Glass fabrics made with E-glass fibres and Camelyaf 266 thermosetting polyester resin (both fibres and resin were obtained from Cam Elyaf Corporation of Turkey) were used to fabricate composites. As an accelerator and initiator for the resin, 0.3 wt.% of cobalt naphthenate (CoNAP) and 1.5 wt.% of methyl ethyl ketone peroxide (MEKP) were used, respectively. The preforming binder was ATLAC 363E, a bisphenol-A based thermoplastic polyester with fumerate groups in the backbone with a melting point of 60 °C.

2.2. Preform consolidation

Preform consolidation was described in detail in the previous study [5]. In brief, glass preforms were consolidated from the glass fabrics by uniformly spreading of 3 and 6 wt.% of the polyester binder onto the glass mats. The binder-coated plies stacked together were pressed under 2.5 kPa and heated for about 45 min at a temperature of 80 °C, which is above the T_m of the binder.

2.3. Preform compaction test

Preform compaction experiments were performed using a Schimadzu^{TM} universal testing machine at a cross head speed of 1 mm/min by applying a compressive load normal to the plane of the fabric preforms placed between compression plates and measuring the distance between the steel circular plates. Preforms composed of eight fabric layers with various binder contents (0, 3, 6 and 9 wt.%) were consolidated based on the procedure described in the previous section. The initial thickness of the preforms prior to compression testing was measured using a micrometer. The measurements were performed carefully without application of any pressure on the samples and average of at least five readings was used in the calculations. The average thickness per layer as a function of compaction pressure was calculated for each preform based on the initial thickness, stroke values, applied force and the area of the preforms, respectively.

2.4. Composite fabrication

Composite fabrication was described in detail in the previous work [5]. As a brief, E-glass fiber/polyester composite plates were manufactured from the consolidated preforms using VARTM process. Preforms were infused with the reacting resin under a vacuum pressure. The applied pressure to the VARTM system by the vacuum pomp was about 10 Pa. After completion of fusion and curing of the resin at room temperature, the cured panels were subjected to post-curing treatment at 110 °C for 2 h. The fiber volume fraction values were measured based on matrix burn-out technique and found to be about 45.3 (±1.05), 50.1 (±1.2) and 46.6 (±0.55)% for the composites with 0, 3 and 6 wt.% binder, respectively.

2.5. Compression test of composites

Compression test method according to ASTM D 695-M was used to measure the ply-lay up and in-plane compressive strength, modulus and strain to failure values of the composite panels with various binder content (0, 3 and 6 wt.%). For this purpose, compression test specimens were sectioned from larger VARTM processed composite panels and tests

along these directions were performed using the mechanical test machine at a crosshead speed of 1.3 mm/min. At least 10 specimens for each set were tested and force versus stroke values were recorded. The compressive stress values were obtained by dividing load values with cross-sectional area of the specimens. The strain was estimated by dividing the adjusted (for machine compliance) stroke values with the initial specimen thickness. The yield stress values were estimated considering the transition values from linear to non-linear behaviour. The modulus values were estimated from the slope of the stress–strain graphs. Failure modes occurred within the specimen during the compressive loading and the effect of the binder on the damage modes were examined using a Philips^{TM} SEM.

2.6. Interactions between binder and matrix resin

Interactions between the thermoplastic binder and the thermosetting polyester matrix were followed to determine the effects of the addition of the preforming binder on the compressive strength and modulus of the polyester matrix and also the extent of the binder dissolution in the reacting resin. For this purpose, 6.5 and 12.85 wt.% of binders (corresponding to 3 and 6 wt.% of the binder in the matrix for 50% fiber volume fraction) were added into the reacting system to replicate the blend of the binder and matrix materials. The blend samples were then subjected to post-curing at 110 °C for 2 h upon room temperature curing. The compressive mechanical properties of the cured model matrix materials were evaluated using the same procedure described in compression testing of the composites. The model specimens were also loaded under flexure and the fractured surfaces of these materials were examined under SEM to follow the level of binder dissolution within the resin. Moreover, the viscosity measurements using a Brookfield LV + rheometer with spindles no. 2 and 3 were performed for neat resin and resin/binder blends. The blends (6.5 and 12.85 wt.% of binder added neat resin) were stirred at room temperature and the viscosity values were recorded in the certain time intervals. In addition, blends with similar compositions were stirred at about 65 °C (above T_m of the binder) for 3 h to simulate the extensive dissolution of the binder by melting.

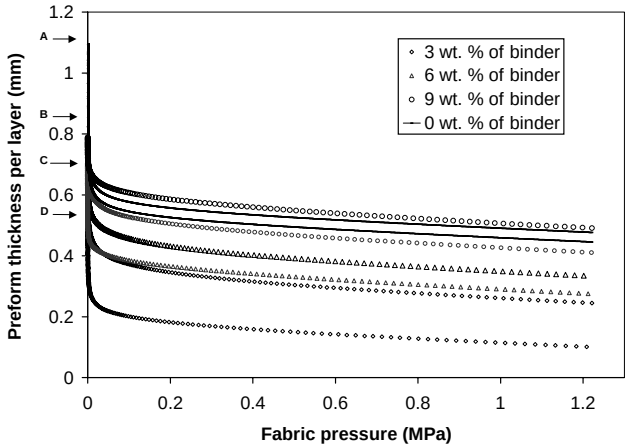


Fig. 1. Preform thickness per layer as a function of fabric pressure for various binder content (A, B, C, D refers to initial thickness of the preforms with 0, 9, 6, and 3 wt.% of binder, respectively).

3. Results and discussion

3.1. Effects of binder on preform compaction

Preform compression tests were performed to evaluate the effect of binder on the preform compaction under compressive loads. This is a particular interest to be considered especially in VARTM processes. Note that the preforms with binder were prepared by stacking the fabric layers together

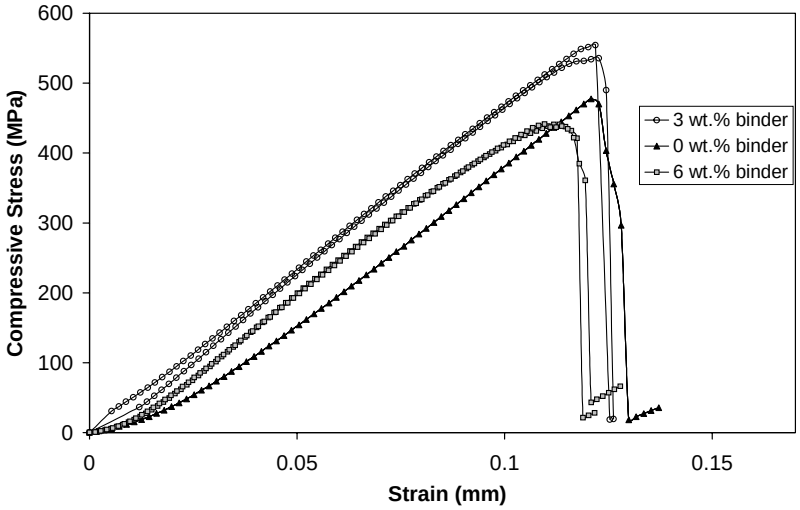


Fig. 2. Compressive stress vs. strain graphs of the E-glass/polyester composite specimens with various preforming binder. The loading was applied in the ply-lay up direction.

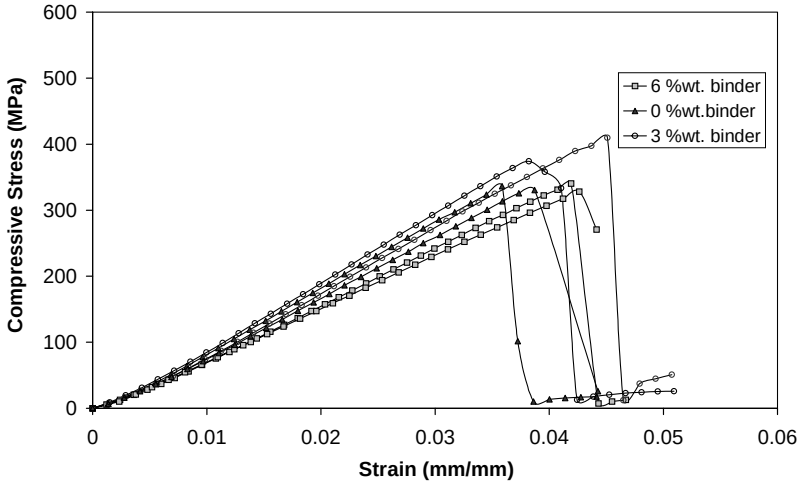


Fig. 3. Compressive stress vs. strain graphs of the E-glass/polyester composite specimens with various preforming binder. The loading was applied in the in-plane direction.

by the application of heat and pressure as described in Section 2.2. The fabric layers were compacted together upon the solidification of the binder particles between the fabrics. Therefore, the thickness of preforms obtained after the thermal consolidation process is much lower as compared to the layered fabrics without any addition of the binder.

Fig. 1 shows the average thickness per layer for the preforms composed of eight fabric layers with 0, 3, 6 and 9 wt.% of the binder as a function of pressure applied by the mechanical test device. The thickness reduction with applied pressure was minimum for the preform with 9 wt.% of binder, while the reduction was maximum for the preforms without binder. In addition, the minimum thickness per layer with applied pressure was achieved with 3 wt.% of binder. Therefore, the highest fiber volume fraction may be expected from this materials under pressure applied during VARTM and RTM processing. As mentioned previously, the volume fraction values of the composites with 3 wt.% of binder were measured to be only about 5% greater than those with 0 and 6 wt.% of the binder. However, the extent of binder dissolution within the reacting resin is a critical issue on the compaction behaviour of the preforms. If the binder dissolves partially during the infusion of the reacting resin prior to the gelation, the binder may be detached from the fiber surface and its compacting effect may be lost and the thickness of the preforms may increase during VARTM. In other words, depend on the type of interaction between the binder and resin, the initial low thickness of the preforms with binder may arise during infusion and reaches to the value of the fabric stacks without the binder. For the material systems studied in this work, partial dissolution of the binder within the resin was observed as mentioned in detail in Section 3.3. So, the detachment of the binder from the fabric surface due to binder dissolution and rise of the prefrom thickness may

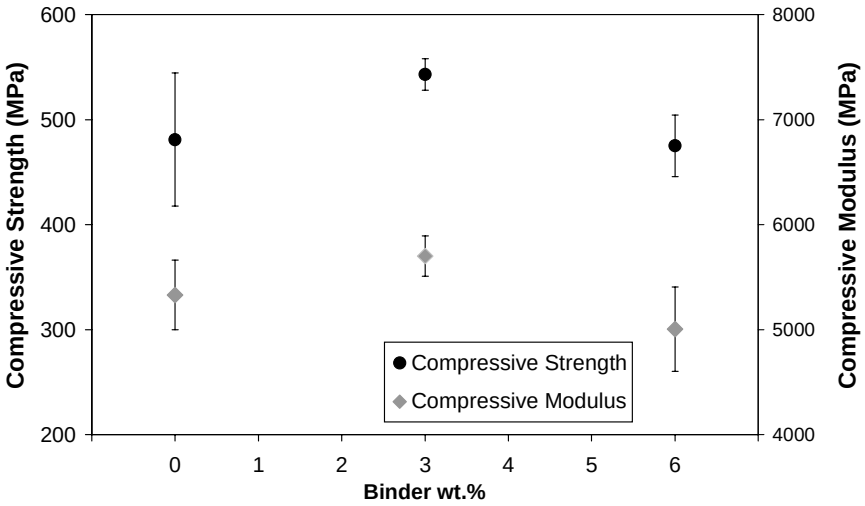


Fig. 4. Compressive strength and modulus values of the composites vs. binder weight percentage for ply-lay up loading direction.

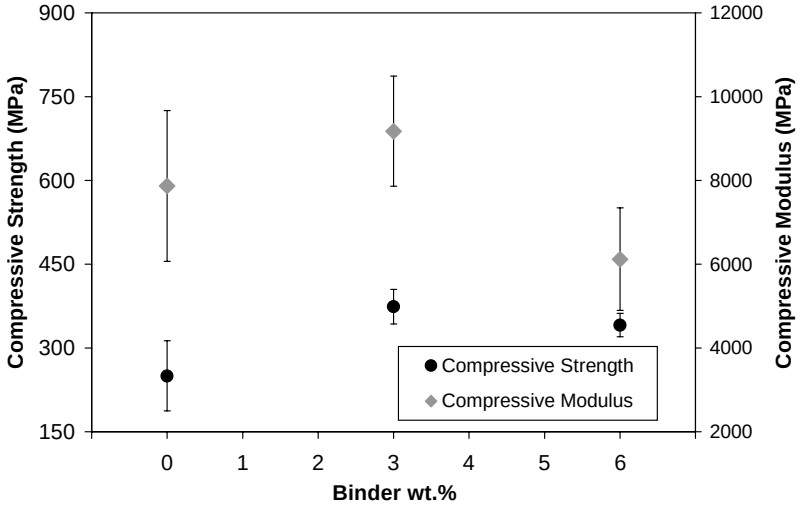


Fig. 5. Compressive strength and modulus values of the composites vs. binder weight percentage for in-plane loading direction.

be expected. In this case, the thickness of the composite panels approaches to each other. The thickness of the composite panels fabricated with and without binder were close to each other (about 13.65, 12.60 and 11.90 for 6, 3 and 0 wt.% binder for the stacks of 25 fabric layers) that indicates the thickness increase of the preforms with binder due to dissolution. Moreover, the peel strength of the E-glass preforms made with polyester powders are in the range of 15–19 N/m as reported in the literature [5]. It was also reported [1] that the peel strength may decreases by up to 80% due to the exposure of the binder to the resin components. Also, note that the vacuum pressures applied during VARTM technique, in general, are relatively low (typically in the range of 0.01–10 kPa) to keep the initial preform thickness. However, the thickness may be kept constant and higher volume fractions may be obtained with binder in the case of application of RTM technique that applies much greater pressures on the fabric layers. These results reveal that application of the binder has some considerable effect on the degree of preform compaction especially prior to resin infusion.

3.2. Compression properties of the composites

Fig. 2 shows compressive stress versus strain responses of E-glass/polyester composites loaded along the ply-lay up direction with 0, 3 and 6 wt.% of binder, respectively. Also, compressive stress versus strain responses of the same type of composites loaded along the in-plane direction are given

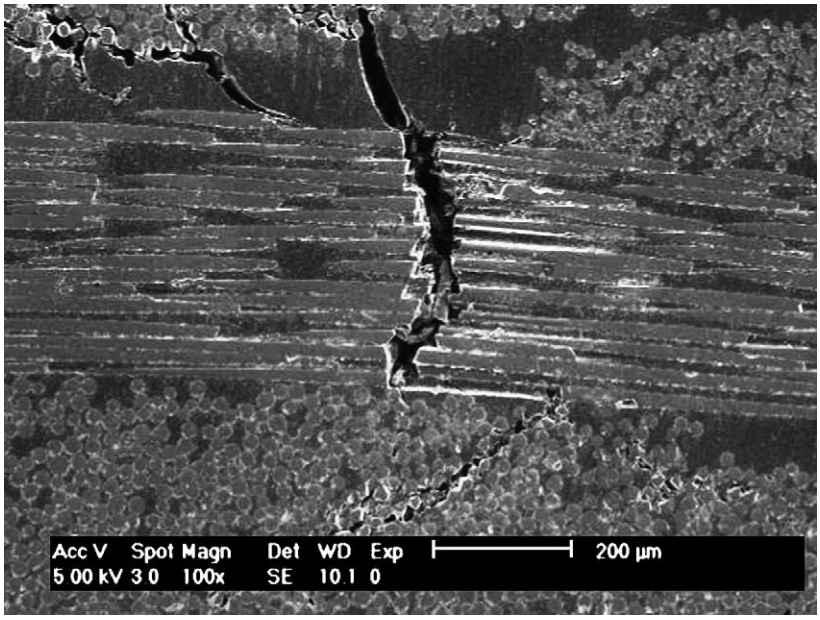


Fig. 6. SEM micrograph showing the fracture modes on the composite specimen without binder loaded in ply-lay up direction.

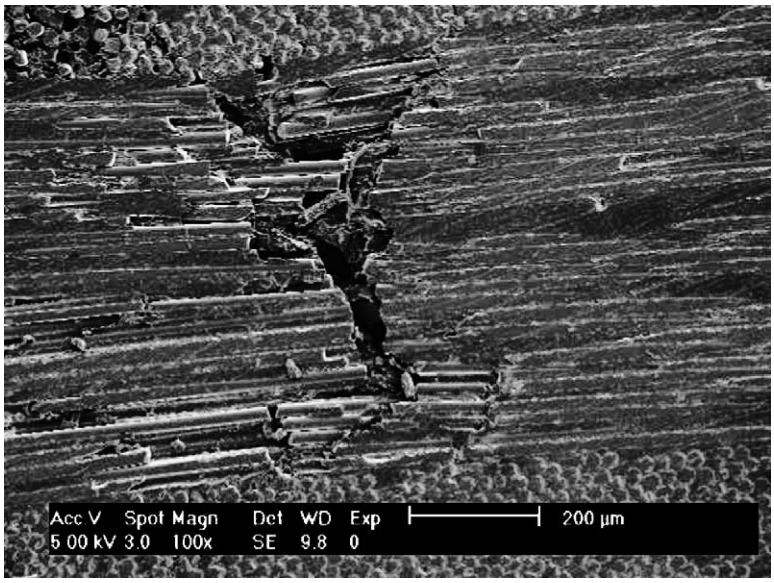


Fig. 7. SEM micrograph showing the fracture modes on the composite specimen with 6 wt.% of binder loaded in ply-lay up direction.

in Fig. 3. For loading in ply-lay up direction, stress–strain response of the composites is almost linear up to the maximum stress level at which damage initiates. There is a sudden drop of the stress after the maximum stress at which failure occurs and the material loses its integrity. The stress–strain behaviour of the composites loaded along the in-plane direction is less linear as compared to those for ply-lay up direction. In some cases, stress increase up to a maximum level without the indication of any damage occurrence. On the other hand, some of the specimens show kinks at stresses prior to maximum level at which the initial cracking occur. The stresses drop suddenly after the maximum stress at which macroscopic damage may occur and material losses its integrity. It was found that the average strain values at maximum stress for the specimens loaded along in-plane and ply-lay up directions are about 0.035 and 0.110, respectively. The results also indicate that the strain values along ply-lay up loading direction are almost constant with the presence of the binder, while those along in-plane loading direction seem to be a little bit increased (about 5%) by introduction of binder. This may be due to the fact that the fracture in in-plane loading is considerably related to the interlaminar properties that are modified by the plastic binder. Figs. 4 and 5 show average compressive strength and modulus values of E-glass/polyester specimens as a function of binder weight percentage for the ply-lay up and in-plane loading

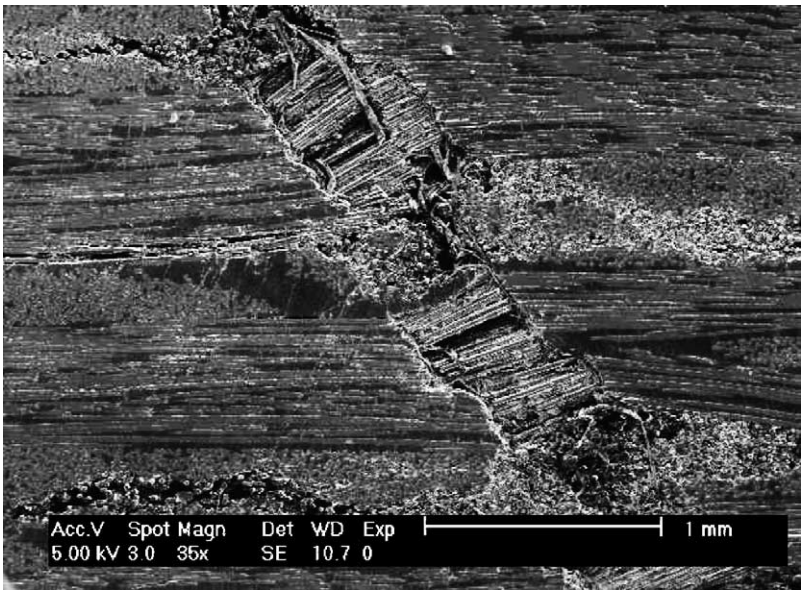


Fig. 8. SEM micrograph showing the fracture modes on the composite specimen without binder loaded along in-plane direction.

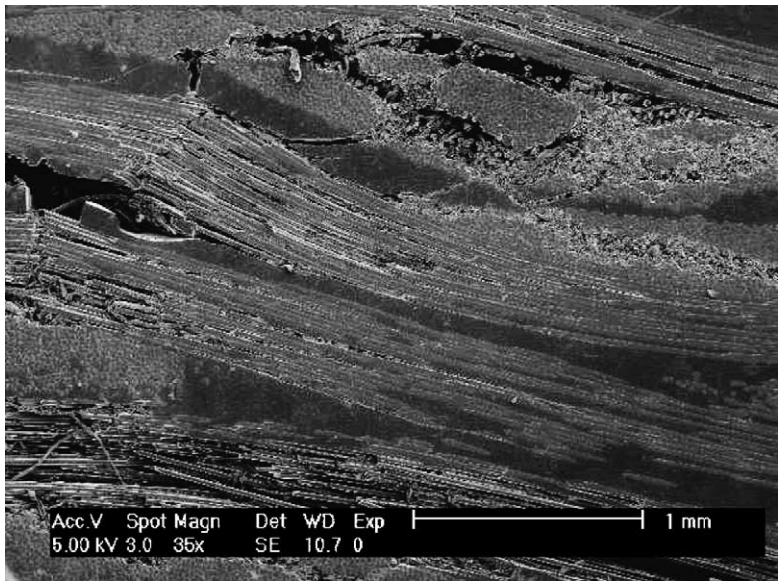


Fig. 9. SEM micrograph showing the fracture modes on the composite specimen with 3 wt.% of the binder loaded in in-plane direction.

directions, respectively. The average strength values are in the range of 400–600 and 150–300 MPa for ply-lay up and in-plane directions, respectively. Also, the modulus values are in the range of 4500–6000 and 5000–11,000 MPa for ply-lay up and in-plane directions, respectively. The higher strength along ply-lay up direction may be due to the fact that the compressive strength is more matrix-property dominant in this direction while along in-plane direction is more related to interlaminar and interfacial bonding. Furthermore, for the both loading directions, both average strength and modulus values increase slightly up to 3 wt.% of the binder while the further addition of the binder results in slight decreases. This may be associated with the optimum interlaminar strength and the highest fiber volume fraction. Fracture surfaces of the fractured compression test specimens for the both loading directions were examined using SEM to reveal

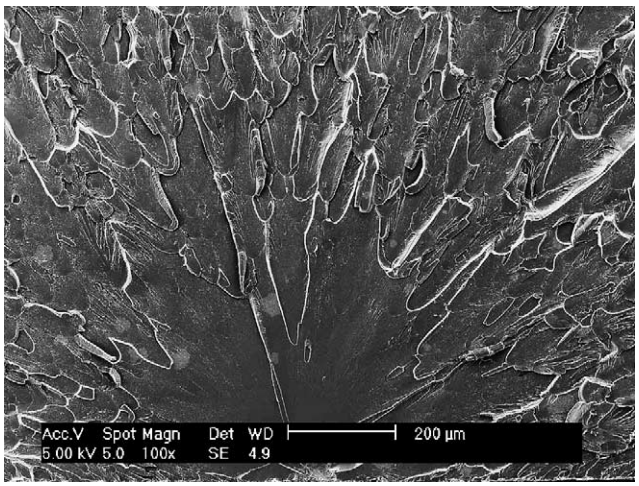
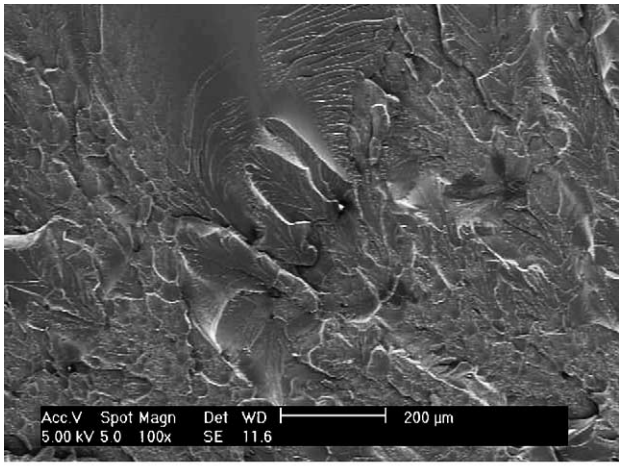
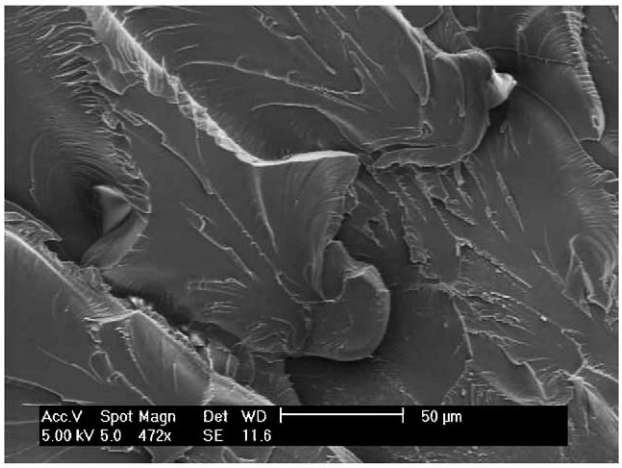


Fig. 10. SEM micrographs of the fracture surface of the cured model matrix material without binder.



(a)



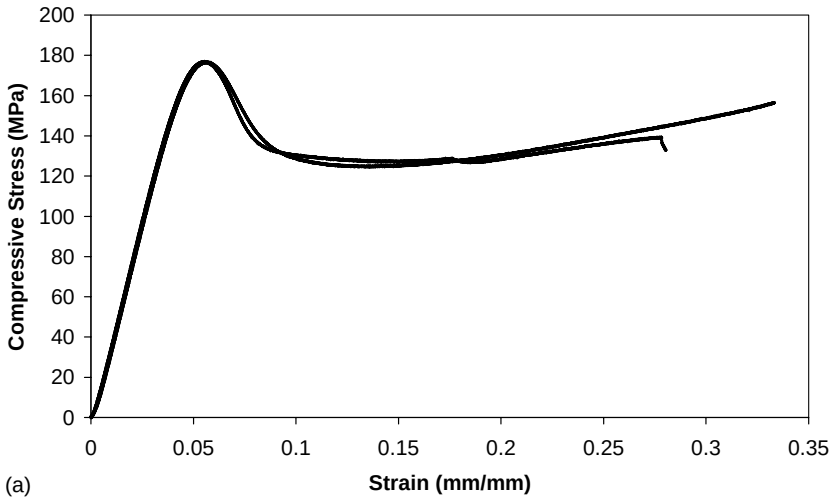
(b)

Fig. 11. SEM micrographs of the fracture surface of the cured model matrix material with 12.85 wt.% of binder (a) at 100× magnification and (b) binder embedded in the matrix resin is visible at higher magnification.

the compressive failure modes at the interlaminar region. Figs. 6 and 7 show the fracture surface SEM micrographs of compression test specimens without binder and with 6 wt.% of the binder for ply-lay up direction, respectively. As seen in the figures, fibres normal to the loading direction were observed to fail due to Poisson expansions. Also, matrix cracking and intrabundle cracks are visible for the specimens with and without binder. Figs. 8 and 9 show the fracture surface of the composite specimens without binder and with 3 wt.% binder for in-plane loading direction. As seen in the figures, fibres along the loading directions were observed to buckle and fractured fibres formed fiber kinks locally. Also, longitudinal splitting, along the interlaminar region is visible. Furthermore, a weaker interfacial bond resulted in intraply splitting and fiber/matrix interfacial debonding. It was also observed that the extend of longitudinal splitting is greater in composites with binder. This may be due to the lower interlaminar strength of the composites made with addition of preforming binder.

3.3. Interactions between binder and matrix resin

Interactions between the thermoplastic binder and the thermosetting polyester matrix were followed to investigate the effects of the binder on the mechanical behaviour of polyester matrix and extent of the binder dissolution within the reacting matrix resin. For this purpose, various amount of binder was added into the reacting system to replicate the matrix material. Figs. 10 and 11 show the fracture surface SEM micrograph of the matrix polymer without binder and with 12.85 wt.% binder, respectively. The specimens were loaded under flexure using three point bending configuration. As seen in Fig. 11b, undissolved binder particles embedded in the matrix are visible. This indicates that there is no complete dissolution of the binder within the reacting resin system. Moreover, as the polyester matrix exhibits a brittle fracture mode (Fig. 10) that is typical for thermosetting polymers, the presence of the binder within the matrix alters the mode of the fracture of the polymer. Mechanical



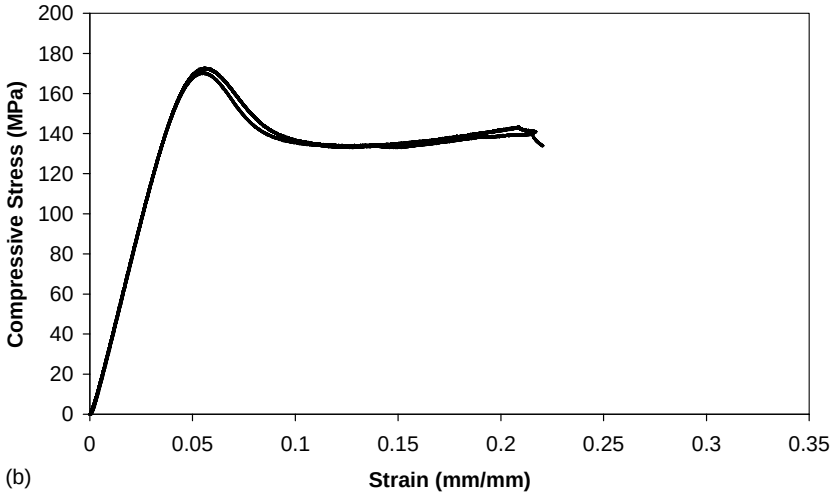


Fig. 12. Typical compressive stress and strain curves of the model matrix material with (a) 0 wt.% binder and (b) 12.85 wt.% binder.

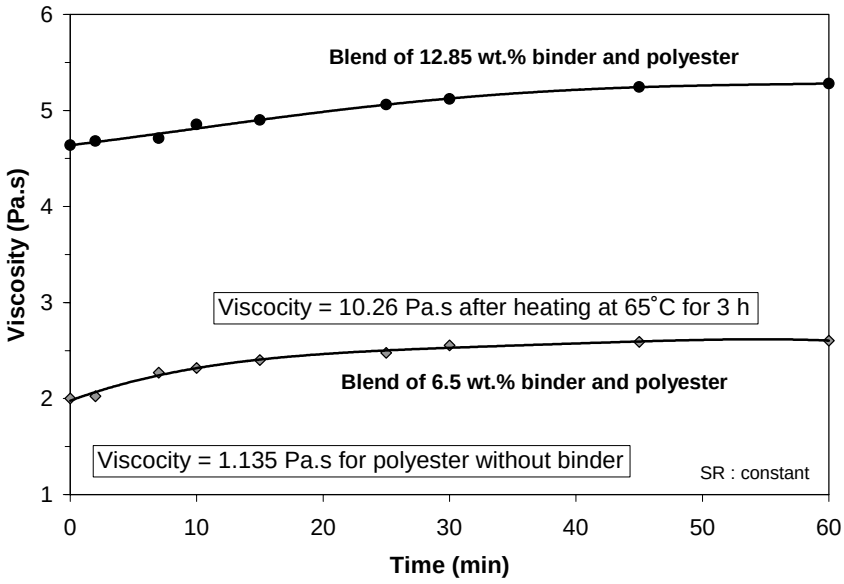


Fig. 13. Viscosity variations of the binder and polyester blend as a function of the time.

behaviour of the cured model materials was also evaluated to reveal the effects of the addition of the binder on the compressive behaviour of the matrix polymer. Fig. 12 exhibits the compressive stress and strain response of the model specimens with and without binder. The stress–strain behaviour of all the specimens showed linear elasticity up to the yield point. The yield point is defined as the point at which deviation was observed in the linear part of the stress–strain curve. For all specimens, deformation involved significant plastic flow beyond the maximum stress and the flow stress remained constant up to relatively high strains. The results reveal that mechanical properties of the model specimens are not significantly affected by the introduction of the binder.

The viscosity values of the resin/binder blends stirred at room and at elevated temperatures are given in Fig. 13. No initiator and accelerator were added to polyester. The viscosity of the polyester without binder and blend after melting of the binder at above the melting temperature of the binder is also given in the same figure. The viscosity of the neat resin at room temperature was measured as 1.135 Pa s. As seen in the figures, the viscosity of the resin lift up to 2.5 and 5.2 Pa s initially by the addition of 6.5 and 12.85 wt.% of the powdered binder. The further increase and then the saturation of the viscosity values were observed due to stirring at room temperature. This further increase may indicate the partial dissolution of the binder within the matrix. The viscosity increase was measured to be about 30 and 15% for 6.5 and 12.85 wt.% of the binder, respectively, up to the gelation time (≈50 min) of the resin in real composites. Moreover, the viscosity value for the blend (6.5 wt.%) increased at elevated temperature (65 °C) and reached to the highest value of 10.26 Pa s at complete dissolution by melting. The results indicate that in real composites partial dissolution of the binder within the resin and therefore an increase in the viscosity of the infusing resin possibly occurs.

4. Conclusion

The present investigation revealed that compressive stress and strain responses of the E-glass/polyester composites loaded along the ply-lay up and in-plane direction were considerably affected by the preforming binder. The effect of presence of the binder on the strain values at maximum stress for both directions were insignificant. For both loading directions, compressive strength and modulus values of the composites increases up to 3 wt.% of the binder and the further increase of the binder concentration results in slight decrease of these values. Furthermore, the preform compaction experiments revealed that the initial fabric compaction can be achieved by application of preform consolidation technique developed in this study using powdered binders and the highest compaction can be obtained with 3 wt.% of the binder. The further addition of the binder results in increasing the thickness of the preforms. However, if the binder dissolves partially within the reacting resin prior to the gelation of the resin, the binder may be detached from the fiber surface and it's compacting effect may be lost and the thickness of the preforms may increase during VARTM. The fracture surface examinations revealed that the failure modes in composites are failure of fibres normal to the loading direction, matrix cracking and intrabundle cracking for ply-lay up direction, and longitudinal splitting along the interlaminar region, intraply splitting, fiber buckling and fiber kink formation for in-plane loading direction. This study also showed that there is partial dissolution of the binder in the reacting polyester resin. Partial dissolution of the binder increases the viscosity of the resin that is critical for the infusion process within the VARTM and RTM techniques. Also, the presence of the binder has some effects on the mechanical behaviour of the matrix polyester matrix.

Acknowledgements

The authors acknowledge the financial support from Izmir Institute of Technology of Turkey under contract 2002 IYTE 06 project.

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