The Influence of Impregnation Condition on Reinforced Ethylene–Vinyl–Acetate Elastomer Composite

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SYNOPSIS

Experiments were conducted to investigate the effect of impregnation conditions on glass fiber-reinforced ethylene–vinyl–acetate elastomer. Both the matrix elastomer resin and reinforcing glass fiber were premixed and compression-molded using a specially constructed mold. The mold prevents the flowing out of the matrix resin during fabrication. The impregnation time was varied between 5 and 25 min. The level of impregnation was measured through the optical micrographs of the cross section, estimation of void contents using the ignition method, and transverse bending strength. The morphology of the fractured surface was studied using scanning electron microscopy (SEM). The results showed that the longer the impregnation time, the lower the void contents. Both the bending modulus and strength increased with increasing impregnation time. The SEM micrograph shows little adhesion between the matrix and the reinforcing glass fiber.

INTRODUCTION

Within the last few decades, several investigations on fiber-reinforced thermoplastic composites have been reported in the literature. These composite materials are enjoying greater attention in both academic and industrial communities due to their greater manufacturing flexibilities and properties. Thermoplastic composites are known to have several advantages which make them suitable for various applications in aerospace, automobile, construction, and other related sectors where high-performance materials are required. In comparison with thermosetting composites, they have unlimited shelf life, damage tolerance, and impact resistance, and the cost of waste is reduced because they can be recycled.[1-4] However, the viscosities of these thermoplastics which are relatively high makes impregnation into the reinforcing fiber a rather difficult task. This problem is receiving wide attention and the last few years have experienced rapid developments in impregnation techniques which include coated yarn, commingled yarn, film stacking, and co-woven fabric.[5-8]

Heretofore, mainly rigid thermoplastic matrix resins but not elastomers were being used in composite fabrication. To correct this oversight was the main objective of this study. In the past, especially in the automobile industry, nonimpregnated fiber-reinforced elastomers were used for making various components, including pneumatic tires and flexible couplings.[9] However, these nonimpregnated composites are difficult to process and often lead to a layer separation. Impregnation can eliminate the above disadvantages.

In this study, the effect of impregnation conditions on the bending properties of glass fiber-reinforced ethylene–vinyl–acetate (EVA) was investigated. The melting point of the EVA (random copolymers) shown in Figure 1 is 91°C.

EXPERIMENTAL

Materials

Pellets of EVA were generously supplied by Toyobo Co. Ltd. The as-received pellets were predried at 80°C for about 12 h. The predried pellets were spun into the fiber using a laboratory-type extruder. The spinneret temperature was 120°C. The glass fiber

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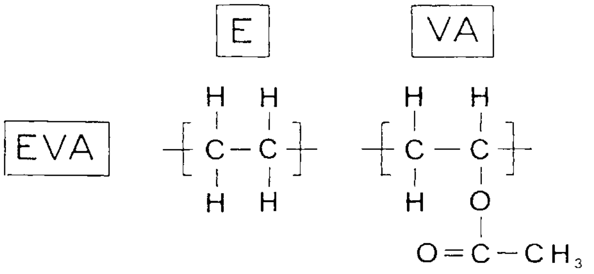

Figure 1 Structure of evathate (E = ethylene, VA = vinyl acetate).

was obtained from Asahi Glass Co. Ltd. The matrix elastomer and the reinforcing glass fiber were unidirectionally premixed to give a volume fraction of 60 : 40. The parallel arrangement of the fiber yarn is represented in Figure 2.

Compression Molding

The compression molding of the premix was carried out by using a specially constructed mold. The main advantage of this mold over the conventional type is that the flow-out of the matrix resin during compression molding is reduced. A full detail of the mold shown in Figure 3 can be found elsewhere.[10] The molding conditions were as follows: mold temperature, 120°C; pressure, 6 MPa; and impregnation time = 5, 10, 15, 20, and 25 min. After molding, the samples were quenched in iced water. To observe the cross section of the samples, small portions were cut along the length of the samples and polished by using various grades of emery paper (240–1500) with a final polishing using a soft cloth with a 0.3 and 0.03 μm diamond paste. The void content was measured by the ignition method. In this case, the matrix resin was burnt off in an oven at 600°C for about 6 h. The void content was computed from the following relationship:

$$\rho_{ct} = \frac{W_c}{\left[\frac{W_f}{\rho_f} + \frac{W_r}{\rho_r}\right]} \tag{1}$$

$$V_v = \left[1 - \frac{\rho_c}{\rho_{ct}}\right] \tag{2}$$

where W_f, W_r, and W_c are the weight fractions of fiber, resin, and composite, respectively; ρ_f, ρ_r, and ρ_{ct}, the corresponding densities; and ρ_{ct}, the theoretical density of the composite.

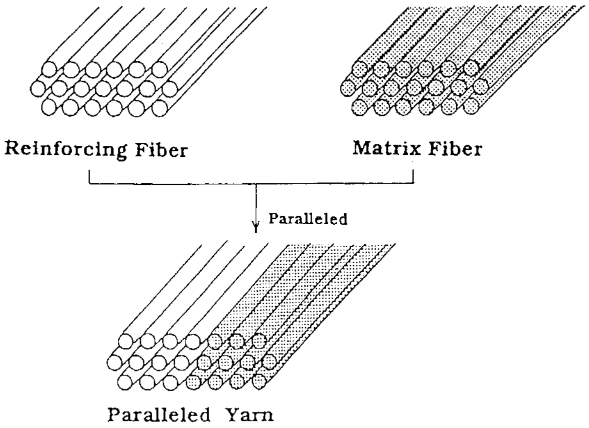

Figure 2 Parallel fiber–yarn arrangement.

Bending Test

The three-point bending test was carried out with a support-to-thickness ratio ("*h*") of 15 : 1. The specimen geometry was 15 h + 20 mm long and 15 mm wide. The test was conducted on batches of three or more samples at room temperature using a Shimadzu Autograph tensile testing machine. The test speed was 5 mm/min, and the investigation was conducted on batches of five in both longitudinal and transverse directions.

Scanning Electron Microscopy

The morphology of the fractured transverse bending samples was examined by scanning electron microscopy using a JEOL Model JSM-5200 at a 15 kV accelerating voltage. The fracture surface of the

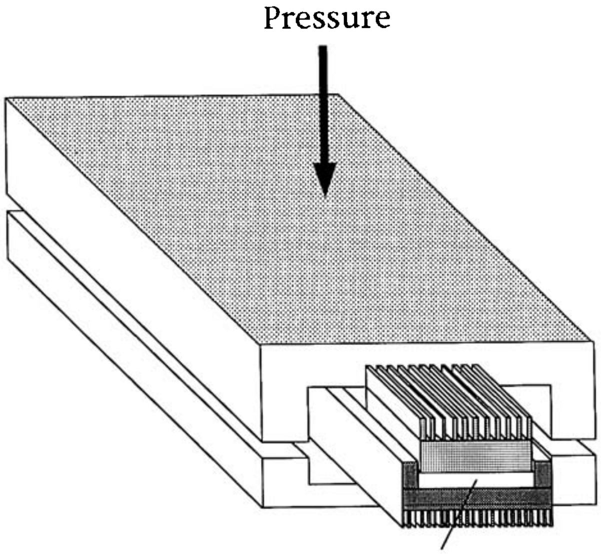

Figure 3 Schematic representation of the mold.

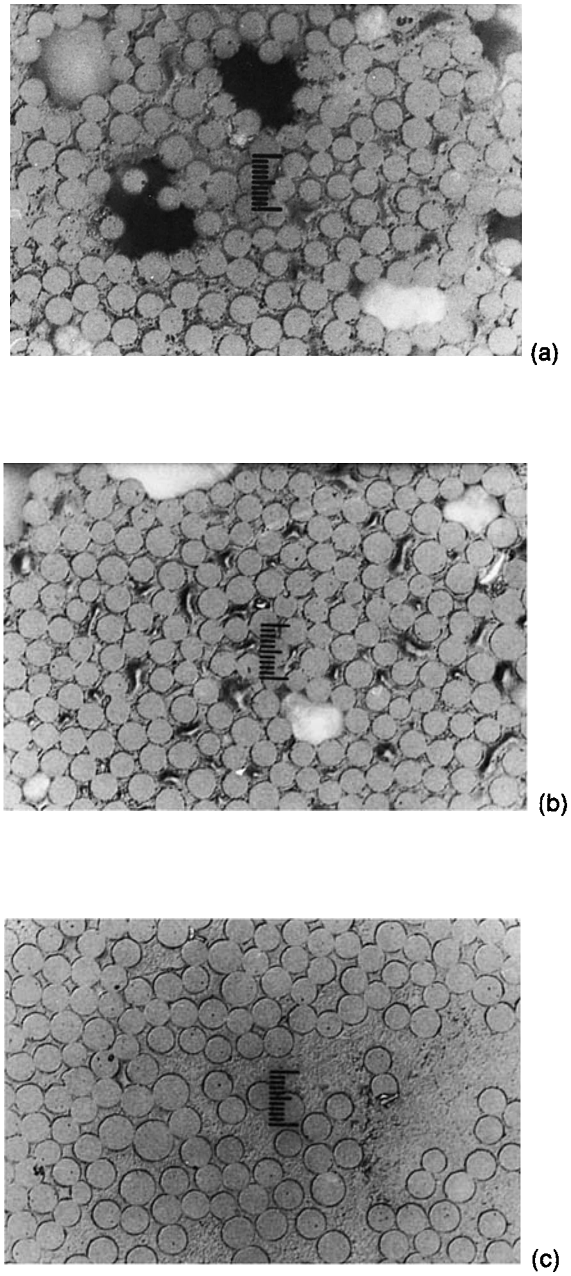

Figure 4 Optical micrographs of cross section of glass fiber-reinforced evathate: (a) 5 min; (b) 15 min; (c) 25 min.

samples was coated with gold and examined at a magnification of 200×.

RESULTS AND DISCUSSION

Degree of Impregnation

To check the impregnation condition of the thermoplastic elastomer composite, the impregnation time was varied between 5 and 25 min. The impregnation conditions were studied through

(a) the observation of voids in the micrographs of cross section,
(b) estimation of void contents, and
(c) transverse bending strength.

The representative micrographs of the cross section of the composite are shown in Figure 4. As can be seen in the figure, the sample impregnated for 5 min contains large quantities of voids characterized by thick dark bubbles (observed under the microscope). The shape and quantities of the voids gradually reduced with increasing impregnation time. In Figure 4(c), where the impregnation time was 25 min, the voids were almost completely eliminated. This situation is confirmed in Figure 5, where the void content reduces with increasing impregnation time. It is seen in Figure 5 that the void content in the 5 min sample was high, i.e., about 8%, and in the 25 min sample, it has reduced to about 2.3%. In fact, the difference in the void content between 5 and 15 min samples was quite small, while between 15 and 20 min impregnation time, a significant reduction in void content occurred. The extent of impregnation can also be understood through the result of the transverse bending strength shown in Figure 6. In this case, the transverse bending strength increases with increasing impregnation time up to 15 min before leveling off. Based on the above observations, it is likely that impregnation of the matrix elastomer resin into the reinforcing glass fiber was complete before 25 min.

The Effect of Void Content on Bending Properties

The presence of voids in the thermoplastic composite can significantly affect the bending properties.

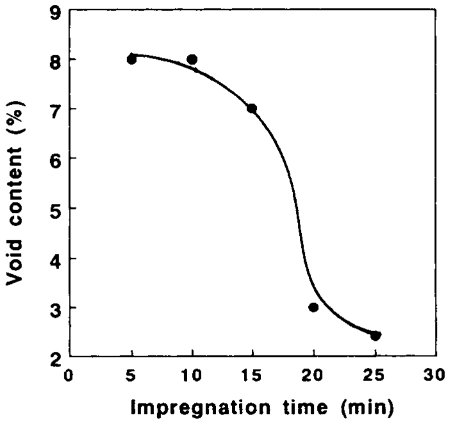

Figure 5 Void content vs. impregnation time.

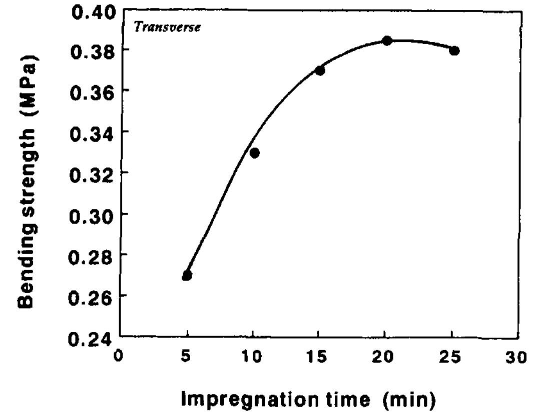

Figure 6 Transverse bending strength as a function of impregnation time.

Thus, a large quantity of voids may lead to a total rejection of the material. Therefore, the main goal of composite manufacturers is to produce void-free materials. It is somehow difficult to achieve a 100% void-free monolithic structure because of the matrix resin which may degrade after a long exposure to a high processing temperature. The effect of void content on the transverse bending strength of glass fiber-reinforced evathate elastomer composite is shown in Figure 7. It is seen in the figure that the sample impregnated for 5 min with a large void content has the lowest bending strength. The high content of voids in the 5 min sample had reduced the bending strength by about 45%. This reduction however, indicates that the longer the impregnation time, the lower the void content and, hence, the higher the bending strength. Further reduction of the void content, i.e., below 2.4%, was difficult to accomplish, because after increasing the impregnation time above 25 min, the sample degraded and the matrix flow-out could not be prevented. Therefore, based on this present experimental technique, the lowest level of void content for a glass fiber-reinforced random copolymer of EVA is about 2.4%. A similar trend also occurred in the transverse bending modulus shown in Figure 8. In this case, the reduction of the bending modulus was more than 100%. The bending modulus is more sensitive to the impregnation time than is the bending strength.

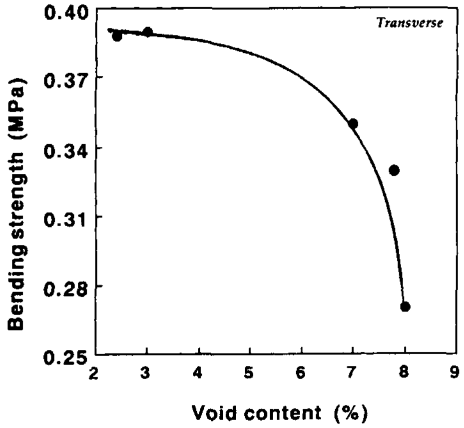

Figure 7 Transverse bending strength as a function of void content.

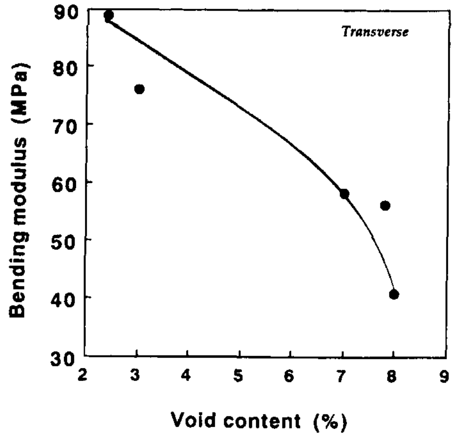

Figure 8 Transverse bending modulus as a function of void content.

Characterization

The bending properties of the reinforced composite are shown in Figure 9. The figure compares the values obtained in 25 min impregnated samples. Longitudinal properties are higher than those of the transverse ones. The bending strength in the longitudinal direction was twice that of the transverse direction. Also, the modulus was more than 50 times higher in the longitudinal direction than in the

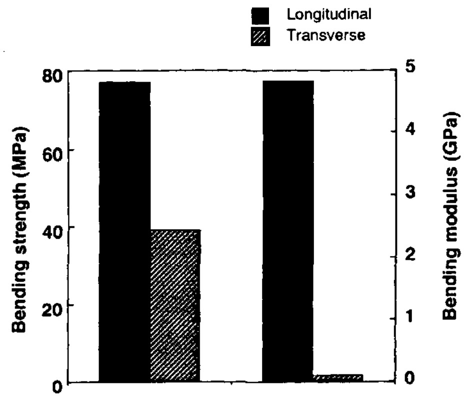

Figure 9 Bending properties at 25 min impregnation time.

transverse direction. High bending properties in the longitudinal direction were influenced by the glass fiber, while the low values in the transverse direction were due to the weak adhesion between the thermoplastic elastomer and the reinforcing fiber. Once again, the modulus is more sensitive to processing conditions than is the strength. The flexible nature of the thermoplastic elastomer was reported by Shonaike and Matsuo[11] to be the major reason why the glass fiber is more effective in modulus than in strength.

Fracture Morphology

The mode of failure of the sample is schematically represented in Figure 10. Normal tensile fracture behavior occurred in all cases. This is in contrast to earlier studies conducted in our laboratory on glass fiber-reinforced poly(ethylene terephthalate).[12] Under identical conditions, reinforced PET (rigid semicrystalline) split along the fiber direction. This indicates that the mode of failure of reinforced thermoplastic composites depends on the type of matrix resin. A representative transverse SEM micrograph of the fractured surface of the elastomer composite is shown in Figure 11. The sample was taken from material with 25 min impregnation time. It is expected that samples impregnated for 25 min will have the best matrix/fiber adhesion. The SEM micrograph in Figure 11 did show little adhesion between the matrix elastomer and the reinforcing glass fiber, i.e., the matrix adhered on the glass fiber. In fact, one would anticipate a better adhesion at the interface but only moderate adhesion was observed. An interesting feature is the matrix reptation caused by the flexible nature of the elastomer.

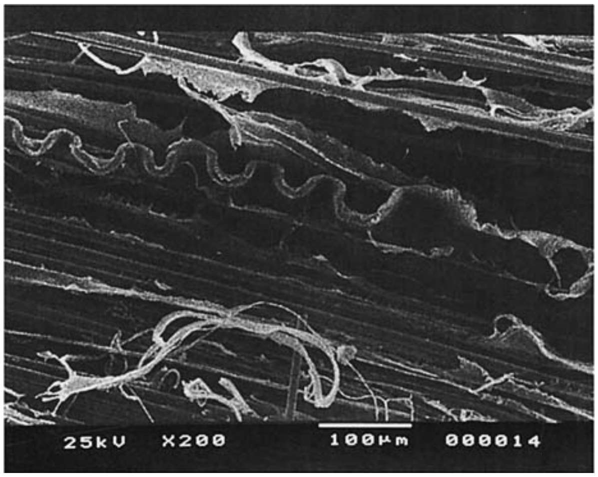

Figure 10 SEM micrograph of fracture surface.

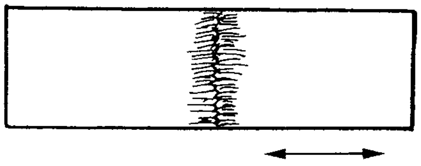

Figure 11 Schematic representation of failure mode.

CONCLUSION

The present investigation has shown the influence of impregnation on bending properties of a glass fiber-reinforced random copolymer of ethylene vinyl acetate. The use of a specially constructed mold reduces the tendency of the matrix resin flowing out during compression molding. The above results showed that the bending properties are strongly dependent on the impregnation conditions. Thus, the longer the impregnation time, the lower the void content and, hence, the higher the bending properties. The SEM micrograph showed little adhesion between the matrix elastomer resin and the reinforcing glass fiber.

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