DEVELOPMENT OF AN ADHESIVE-PRIMER FOR POLYPROPYLENE COMPOSITES

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Abstract

Joining is often one of the critical steps in the fabrication of composites products. However, the low polarity and inert characteristics of polypropylene composite surfaces cause many problems in the assembly of these composites with dissimilar materials. In order to overcome the adhesion issues, an epoxy based primer was developed and the compatibility of several commercial adhesives with the primer was evaluated. Results showed very good lap-shear strength of up to 15 MPa with substrate failure. The performance of the primer was also evaluated between -30 and 80°C and after conditioning in humid condition. While the lap-shear strength decreased with increasing temperature, it remained unchanged after conditioning. Finally, different practical approaches to apply the primer film to a polypropylene continuous fiber composite were investigated, including techniques to apply the primer during and after composite consolidation.

Introduction

Polymers such as polypropylene are difficult to bond because of their non-polar and inert surfaces. One successful approach to overcome this difficulty involves proper surface preparation prior to bonding. Surface preparation techniques generally involve the introduction of functional groups at the surface in order to promote bonding between the substrate and the adhesive. Surface preparation techniques that are widely accepted include chemical etching and plasma, flame, or laser treatments. Another successful approach to bond non-polar and inert polymers can involve the use hot-melt adhesives. However, heat-gun techniques for hot-melt adhesives are often not sufficient for joining large areas while surface preparation techniques may not always be practical in industrial applications. When this is the case, primers can be alternatively used. Primers are applied to part surfaces prior to bonding in order to introduce reactive groups. Primers thus form a chemical bridge between substrate and adhesive. Several studies can be found in the literature on the development of primer specifically formulated to improve the adhesion to polypropylene or polyolefin's [1-3]. However, there remains a need in the industry for an effective and easy-to-use primer for use in the repair, assembly and/or coating of polypropylene composite materials, especially with dissimilar materials like, for example, thermosets and their composites, thermoplastics and their composites, metals, wood, etc. The availability of such a primer would help solve some of the adhesion issues that have significantly restricted the introduction of polypropylene composite material in several fields of applications.

In this study, the performance of a newly developed primer is evaluated. To achieve this objective, the primer compatibility with several commercial adhesives was first evaluated. Then, the influence of environmental factors on the performance of the adhesive-primer was measured by performing tests at different temperatures and after conditioning in water and in a hot/wet environment. Finally, practical approaches to apply the primer film to a polypropylene continuous fiber composite were investigated, including techniques to apply the primer during and after composite consolidation.

Experimental

Primer Formulation

Epoxy-functionalized polypropylene primer was prepared in a twin-screw extruder by mixing maleic anhydride grafted polypropylene with an epoxy resin at a temperature of 220°C [4]. During extrusion, the epoxy resin reacted with the functional group of the grafted polyolefin to create a primer which composition comprises the reaction product of the reactive epoxy with the grafted polyolefin. A 10 μm film of the prepared primer formulation was then produced by calendaring using a material temperature of 200°C.

Material and Mechanical Characterization

The glass fiber reinforced polypropylene composite studied in this research consisted in a continuous fiber fabric woven into balanced 2-2 twill having nominal weight of 1485 g/m^2. Fabric had a fiber weight fraction of 60%, giving a final consolidated fiber volume fraction of 35%. To prepare plates, three layers of fabric were superposed in a flat closed-mold. Consolidation cycle then consisted in heating the fabrics in a closed-mold to a temperature of 200°C, holding this temperature for five minutes, and then cooling the fabric at a rate of 10°C/min. A pressure of 0.7 MPa was applied during the entire molding cycle. Unless otherwise stated, the primer film was fused on top of composite coupons using a small heated press whereas only the upper plateau was pre-heated to a temperature of 220°C. Prior to inserting the materials into the press, a thin release film was put on top of the primer surface to avoid having the primer stick to the upper plateau. Pressure was then applied for a few seconds until the adhesive-primer was completely melted to the composite surface. Once this was achieved, pressure was removed and material was cooled at ambient temperature (23°C).

To evaluate the performance of the newly-developed adhesive, lap-shear strength was evaluated on specimens made of overlapping aluminum and glass fiber reinforced polypropylene composite coupons (Figure 1). Coupon dimensions were of 10 x 2.5 cm overlapped by 1.25 cm. Prior to bonding, aluminum coupons were sandblasted and acetone washed to ensure surface cleanliness. Sandblasting was performed a maximum of 4 hours prior to bonding. The compatibility of several commercial adhesives with the primer was evaluated by performing tests with three different types of commercial adhesives, i.e.:

- An epoxy adhesive (Hysol® 9340);
- A methacrylate adhesive (Araldite® 2022);
- A polyurethane adhesive (Araldite® 2041).

Adhesives were cured at room temperature for a minimum of 15 hours before being post-cured 2 hours at 60°C. To maintain alignment, a small pressure of 12 psi was applied on the overlapping area of the test specimens for the entire curing and post-curing period. Specimens were also prepared by laminating a glass/epoxy composite on top of the polypropylene composite to which a primer film was previously fused. The glass/epoxy composite consisted in an epoxy resin laminated with three layers of 22 oz/yd2 fabric for a total approximate thickness of 3 mm.

Lap-shear strength was also determined at temperatures ranging from -30 to 80°C using an environmental chamber mounted on an Instron universal testing machine and at room temperature (23°C) on specimens previously conditioned in water and in hot/humid environment (60°C and 80% humidity) in order the influence of environmental factors on the performance of the adhesive-primer.

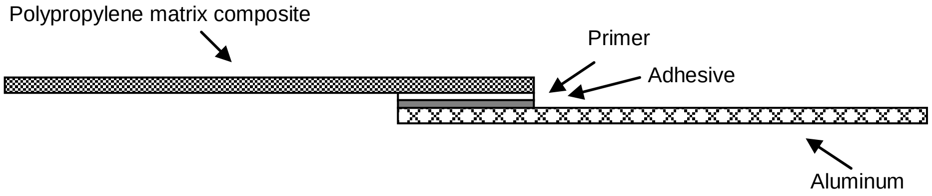


Figure 1: Specimen configuration used for lap-shear strength experiments.

Results and Analysis

Compatibility with Commercial Adhesives

The compatibility of the newly developed adhesive-primer with commercial adhesives was evaluated by performing lap-shear tests on specimens prepared with common adhesives, i.e. an epoxy adhesive, a methacrylate adhesive and a polyurethane adhesive. As benchmark, lap-shear specimens were also prepared without primer using the epoxy adhesive. As expected, lap-shear specimens prepared without primer are poorly bonded and can thus be easily separated by hand. Lap-shear tests gave strength of only 2.3 MPa (Figure 2). The use of primer provided very significant improvement in adhesion. For epoxy adhesive, lap-shear strength of 15 MPa was obtained using the developed primer. As shown in Figure 3a, failure occurred both in the adhesive and in the composite, which provides evidence of good adhesion between the primer and the adhesive. In certain areas, resin of the composite was completely peeled-off from the composite (substrate failure), leaving un-impregnated fibers on the surface of the composite. In some other case, yarns were partially peeled-of from the composite surface. For urethane adhesive, lap-shear strength of 7.8 MPa was obtained. As shown in Figure 3b, failure occurred at the primer-urethane adhesive interface. Results obtained for the methacrylate adhesive are much lower than the previous ones. For this adhesive, lap-shear strength of 2.2 MPa was obtained. Observation shows failure at the primer-adhesive interface, indicating poor compatibility between the primer and the adhesive (Figure 3c). Finally, results obtained for the laminated glass epoxy composites were found to be very similar to those obtained for the epoxy adhesive, since results showed lap-shear strength of 14 MPa. Inspection of the failure surface showed once again substrate failure of the polypropylene composite, the polypropylene matrix being peeled-off the composite surface. These results clearly indicate that the best compatibility with the primer is achieved with epoxy.

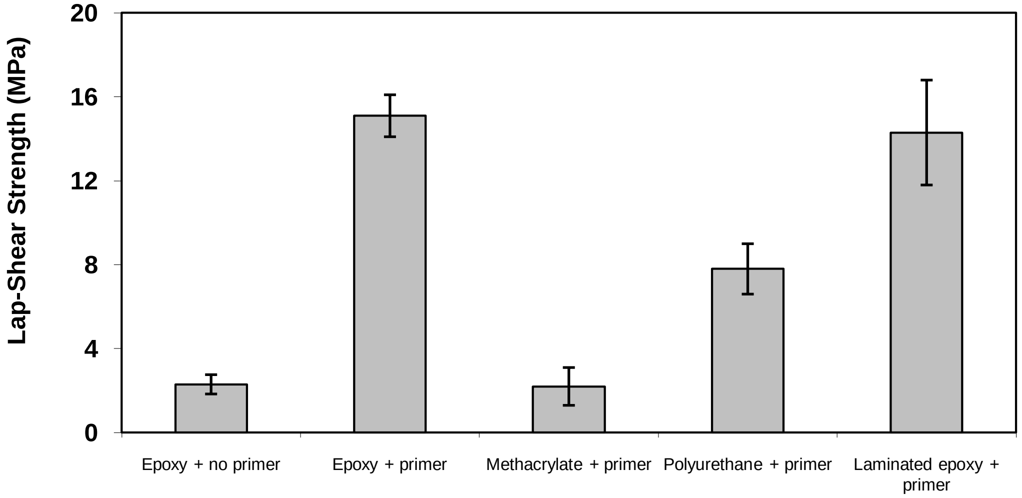


Figure 2: Lap shear strength obtained at 23°C using several commercial adhesives and a laminated composite.


(a) (b) (c)

Figure 3: Surface failure obtained for polypropylene/aluminum lap-shear specimens fabricated using the developed adhesive-primer and (a) an epoxy adhesive (b) a urethane adhesive and (c) a methacrylatye adhesive.

Conditioning

To measure the influence of environmental factors on the performance of the adhesive-primer, two series of tests were conducted. In the first series, the performance of the primer was evaluated at temperatures ranging from -30 to 80°C. In the second series, the performance of the adhesive was measured after immersion in water and after exposition to a hot/humid environment for a period of up to 30 days. These tests were all performed using specimens prepared with the primer and the epoxy adhesive, since this combination yielded the best results in the previous section.

Variation of the lap-shear strength as a function of the temperature is shown in Figure 4. As shown in this Figure, lap-shear strength decreases with increasing temperature following a linear trend. Under the range of temperature investigated, the lap-shear strength vary significantly from approximately 20 MPa at -30°C to only 6.8 MPa at 80°C. Observation of the failure surface indicated change in failure mode with temperature. For temperature of 60°C and below, a combination of substrate and adhesive failure similar to the one shown in Figure 3a was observed. For a temperature of 80°C, failure occurred in the adhesive. Reduction in lap-shear strength and change in failure may be attributed to the softening of the polypropylene and adhesive with increasing temperature. Results of the tests obtained on specimens conditioned in water and by and in hot/humid environment are shown in Figure 5. As shown in Figure 5a, conditioning in water did not resulted in a loss in mechanical performance even after a 30 days immersion. Observation of the failure surface showed a combination of substrate failure and adhesive failure similar to the one shown above for dry specimen (Figure 3a). Conditioning at 60°C in an environment containing 98% of humidity yielded similar results since no loss of mechanical properties was observed. Failure mode observed was also a mix of substrate failure and adhesive failure.

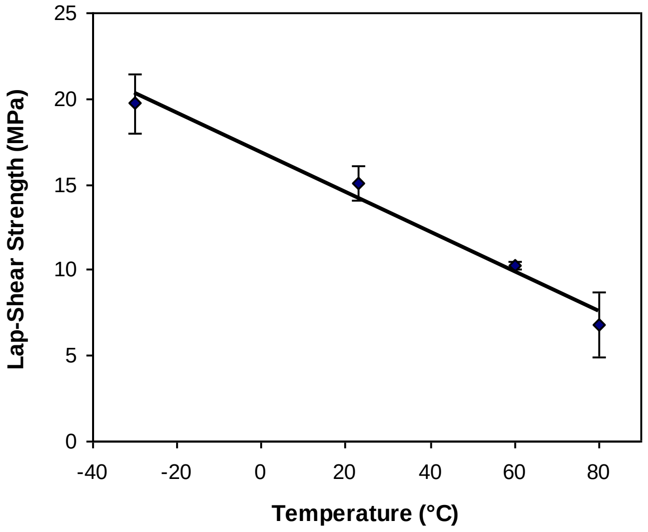
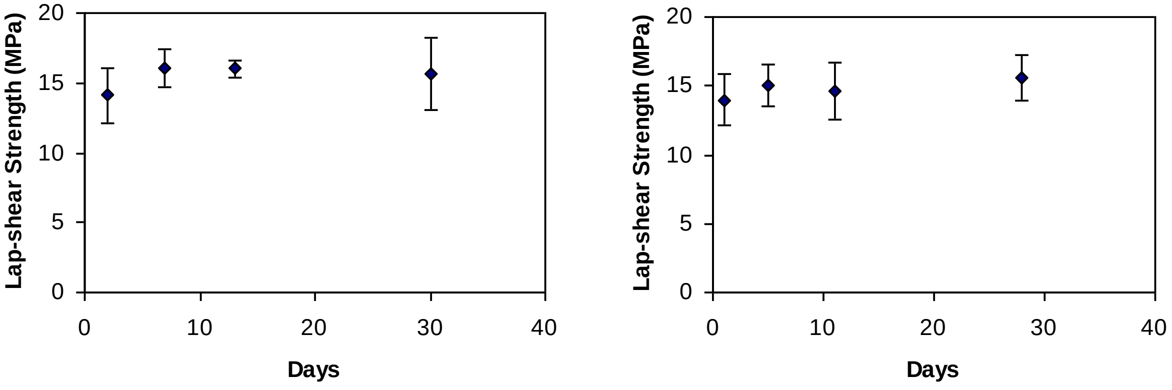


Figure 4: Lap shear strength measured as a function of temperature.



(a)

(b)

Figure 5: Lap shear strength measured on (a) specimens conditioned in water and (b) conditioned in hot/humid environment.

Practical Approach to Apply the Primer

Two different approaches were investigated to apply the primer film to the polypropylene matrix composites. The first approach investigated consisted in bonding the adhesive-primer to a comingled polypropylene matrix composite during vacuum forming. To achieve this, one layer of adhesive-primer was first lay-down in the bottom of a mold. Unconsolidated composite plies were then superposed on top of the primer and later enveloped by vacuum films, as in the case of thermoset composites. Once under vacuum, the assembly was then heated to a temperature of 200°C in a convection oven to allow the thermoplastic matrix to impregnate the glass fibers. After cooling, the composite part was de-molded. Although vacuum forming is generally limited to small series of production, this process offers in the present case the advantage of adding the primer during the processing stage of the composite, thus reducing the necessary number of steps to bond a dissimilar material to the polypropylene composite. The second approach investigated consisted in applying the primer film to the composite surface with the use of a heat source and a small roller (Figure 6), similarly to what can be done in tape placement or film stacking (5). During this operation, the air temperature exiting from the heat source and the displacement speed were adjusted in order to allow the surface of the composite and the surface of the adhesive-primer to melt and then later to solidify under the compression force applied by the roller. Following both approaches, the procedure described above to prepare lap-shear specimens with an epoxy adhesive was used.

Results obtained with both techniques are summarized in Table 1, along with the results obtained previously by applying the adhesive-primer by compression molding. For the vacuum forming process, lap-shear strength of 11.5 MPa was obtained. This is slightly lower than what was obtained by adding the primer to the composites with a small press with heated plateaus, i.e by compression molding. A possible reason for that reduction in mechanical properties could be the migration of polypropylene matrix at the primer location during vacuum forming. Such migration of polypropylene matrix can be expected to result in a reduction of the adhesion. Results obtained for the application of the adhesive-primer with a heat source and a small roller are significantly lower to those previously obtained, as lap-shear strength of only 2.5 MPa was measured. The reduction in lap-shear strength is thought to be the result of the non-uniform heating and pressure obtained as the heat source and roller are moved manually. Further modifications will be performed on the experimental set-up in order to optimize the primer application.

Figure 6: Application of the adhesive-primer using a heat source and a small roller.

Table 1: Influence of application process on the lap-shear strength.

Application process	Lap-shear strength (MPa)
Vacuum forming	11.5
Heat source and roller	2.5
Compression molding	15.0

Conclusions

An epoxy-based primer was developed to improve the adhesion of polypropylene matrix composites to dissimilar materials. First, the compatibility of the adhesive-primer with different adhesives was evaluated. Results showed that very good lap-shear strength of 15 MPa with substrate failure can be obtained when bonding a polypropylene matrix composite to aluminum using an epoxy adhesive. The performance of the primer was also evaluated between -30 and 80°C and after conditioning in humid condition. While lap-shear strength decreased with increasing temperature, it remained unchanged after conditioning. Finally, different techniques to apply the newly developed primer to a polypropylene composite were studied. This revealed that satisfactory results can be achieved by compression molding and vacuum forming process.

References

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