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Impact properties of layer by layer in-situ UV cured composites

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Abstract

Fast and eco-friendly composite curing technologies are highly sought after, and ultraviolet (UV) curing offers both features. UV curing can be applied as a bulk curing technology as long as light arrives at all the points in the material with enough intensity. Additionally, the outer layer cures almost instantaneously, reducing the emission of volatile organic compounds. Despite its benefits, the implementation of UV curing technology in composites manufacturing is still in an early stage, and more basic knowledge about its principles needs to be generated. For instance, when curing considerably thick laminates, the light arriving at the points in the material further from the exposed surface is insufficient for a one shot consolidation, requiring a layer-by-layer curing strategy.

In the present paper, we explore several approaches for the layer-by-layer curing of a glass fibre reinforced unsaturated polyester composite. We assessed the impact properties of the manufactured laminates through drop weight low velocity impact tests to quantitatively evaluate the different approaches. We found that the laminate cured in three steps, applying four plies by shot, had the best impact performance, followed by the composite cured in two steps. On the other hand, the laminate cured in one shot had the worst impact properties, dissipating 20% less energy than the best performing laminate. Therefore, we conclude that the bonding of the UV cured laminates is tough and that the residual stresses induced during manufacturing are lessened the thinner the layers cured in each step are.

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1. Introduction

The broader industrial use of composite materials is limited by their high operational costs and the complexity of the currently employed manufacturing technologies. Considering that almost all the industrial applications of composites rely on thermal curing to consolidate parts, the development of processes based on other curing technologies is a promising option to outperform the state of the art.

In addition to thermal contact, electromagnetic radiation is another way of delivering enough energy to trigger the polymerization of the resin system in a composite. Inside the electromagnetic spectrum, infrared [1], microwave [2], or ultraviolet (UV) radiations [3] have been successfully employed for curing composites. By substituting the thermal catalyst from thermoset resin formulations such as vinyl ester [4], epoxy [5] and polyester [6] with a photoinitiator, composites can be cured in a matter of minutes, or even seconds, through exposure to UV light. And since the basics of the resin formulations are maintained, the mechanical properties achieved do

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not differ from the ones obtained in their thermally cured counterparts. In addition to the benefits of an increased curing velocity, the emission of volatile organic compounds (VOCs) from the resin polymerization is reduced because the UV curing reaction propagates from the irradiated surface to the inside of the parts [4]. The unlimited pot life of UV curable resins implies that fibre reinforcement can be impregnated without worrying about premature gelation, and resin excess can be collected for reuse, reducing scrap. However, only composites reinforced with materials transparent to UV light, like glass fibre, can be cured by these means [7]. The penetration of UV radiation in the material is the main limiting factor for the technology, allowing to efficiently cure laminates with a maximum thickness of between 8 and 13 mm [3], depending on the capacity of the UV sources and the effectivity of the resin formulation employed. But this constraint can be overcome by implementing UV curing in processes in which the composite does not have to be cured as a bulk, like automated tape laying (ATL), automated fibre placement (AFP) or filament winding, where thin layers could be successively placed, and *in-situ* UV cured.

The interlaminar shear strength and fracture toughness of the UV cured secondary bonded composites are similar to thermally cured ones [4], [8], [9]. However, the effect of the processing route of such layer by layer *in-situ* UV cured composites on their mechanical functionality has not been thoroughly studied in the literature. Therefore, the present paper assesses the effect that this layer by layer approach could have on the final mechanical properties and on the impact resistance of composite parts. These points have been addressed by comparing the bending and drop-weight impact behaviour of bidirectional glass-fibre/polyester UV cured composites manufactured following three different layer by layer strategies.

2. Methodology

2.1. Materials

The composite used in this study was a glass fibre/UV cured polyester. The UV curable resin formulation was an unsaturated polyester (FPC-7621 NA, Irurena S.A.) to which a photoinitiator system had been added (a combination of Irgacure 379 and Irgacure 819). The reinforcement was an E-glass fibre fabric with plain weave architecture and a 300 g/m2 surface weight. All the manufactured laminates were reinforced with 12 plies.

2.2. Composite processing

Flat plates of 250 x 300 x 3 mm were obtained by infusion using a 6 mm thick quartz glass as a mould. The laminates were irradiated through this quartz glass with a DYMAX 2000-PC flood lamp with a 75 mW/cm^2 iron bulb.

Three different strategies were adopted to UV cure the composite. The first laminate, identified as UV12, was made of 12 glass fibre plies polymerized in one single shot after a 10 minute exposure to UV light. The second laminate (UV6) was manufactured in two steps. First, six plies were laid and UV cured, and then another six more were laid over the initial ones and UV cured for another 10 minutes, resulting in a secondary bond between the 6th and the 7th plies. The third UV cured laminate (UV4) was manufactured in three steps, laying and curing four plies each time. As a result of this process, the UV4 composite had two secondary bonds, one between the 4th and 5th plies and the other between the 8th and 9th.

2.3. Physical properties

The density of the composite was determined according to ASTM D792-08, while the fibre volume fraction was measured following the procedure described in ASTM D3171-09.

2.4. Flexural response

Three point bending quasi-static tests were performed according to ASTM D790-10. Tested samples were 12.7 mm wide and 3 mm thick. A span to thickness ratio of 16 to 1 was set, and tests were carried out at 1 mm/min in a universal mechanical testing machine equipped with a 5 kN load cell. Samples deformation was measured with strain gauges, and a minimum of five samples were tested for each of the adopted manufacturing strategies.

2.5. Impact behaviour

Low-velocity impact tests were conducted in a drop-weight testing machine (Fractovis- Plus, Ceast). This machine records the contact force history during impacts using a 20 kN load cell attached to the striker and is equipped with an antirebound device to ensure that the striker hits the sample only once during each test. The striker's head had a 20 mm diameter hemispherical geometry for impacting on the 60 mm diameter circular samples that were fixed to the testing setup using a clamping ring with an inner diameter of 40 mm and an outer diameter of 60 mm. The impact energies applied ranged from 0.6 J to 40 J and were defined by setting the striker mass to 2.045 kg and modifying the initial height of the striker to get impact velocities ranging from 0.98 m/s to 4.43 m/s. At least three samples were tested for each impact energy.

3. Results

3.1. Density and fibre volume fraction

The density and fibre volume fraction of each composite are reported in Table 1. The slight differences obtained could be related to the variability of the infusion process itself more than to the different curing strategies adopted. Ultrasonic C-scans were completed on the laminates manufactured by the proposed UV curing strategies, and as a result, no delaminations nor significant size porosities were found.

	Fibre content in volume (%)	Density (g/cm^3)
UV12	50.4 ± 0.2	1.89 ± 0.05
UV6	46.9 ± 0.1	1.85 ± 0.06
UV4	45.7 ± 0.1	1.83 ± 0.05

Table 1. Physical properties of the studied laminates.

3.2. Flexural response

The feasibility of the proposed layer by layer *in-situ* UV curing strategies is conditioned to the mechanical properties achieved on the manufactured laminates. The bending performance of all the UV cured laminates was alike, and in the three cases, the brittle failure behaviour that could be expected was observed. The flexural stiffness and strength of the three laminates are reported in Table 2. Specific stiffness and strength are also reported since fibre volume content and density were slightly different between laminates.

	E (GPa)	σ_{max} (MPa)	$E^{1/2}/\rho$	$\sigma_{\text{max}}^{2/3}/\rho$
UV12	16.25	441	2.13	30.65
UV6	18.84	476	2.35	32.95
UV4	19.17	463	2.39	32.70

Table 2. Elastic modulus (E), flexural strength (σ_{max}) and their corresponding specific values for the studied laminates.

The secondary bonds present in the laminates manufactured following a stepped strategy, that is to say, UV6 and UV4, did not affect their bending performance. These two laminates had almost the same flexural properties, whereas those from the UV12 laminate cured in one shot were somewhat worse. This outcome may be related to the higher temperatures reached during the bulk UV curing of the UV12 laminate since more resin reacts at once, generating more residual stress in the composite.

3.3. Impact behaviour

The impact behaviour of composites is a complex phenomenon with many factors involved. However, it provides descriptive and quantitative information to make a comparative analysis of different composites. Impact resistance can be studied by looking at the evolution of the maximum peak load withstood by the material with the impact energy or by determining the penetration energy threshold at which all the impact energy is dissipated.

The peak load curves of the three laminates (Figure 1) show a typical evolution for thin composite plates. For the lowest impact energies at which only delamination is experienced by the samples, the peak force recorded during impact tests increases with the incident impact energy. But when other damage mechanisms, such as fibre breakage, are initiated, the peak load tends to reach a plateau in correspondence with the Mean Static Ultimate Load. All laminates had a similar maximum load, which is consistent with the flexural test results.

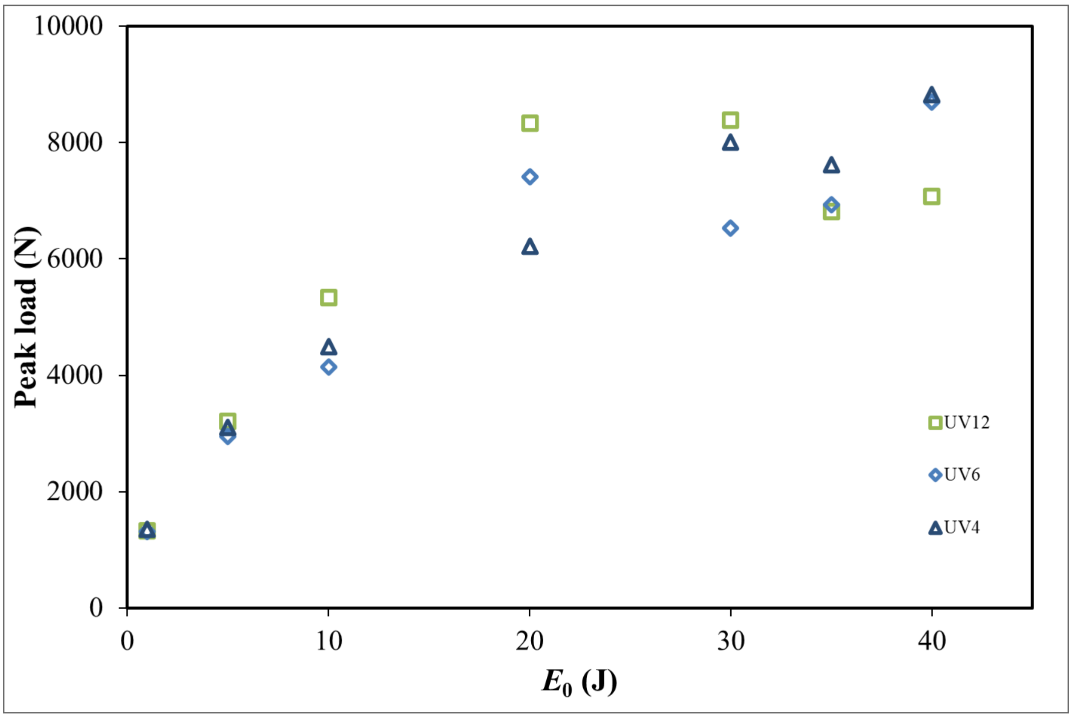

Figure 1. Peak force as a function of impact energy (E_0) for the studied laminates.

Figure 2 shows the dissipated impact energy (E_{dis}) registered in the low velocity impact tests as a function of the incident impact energy (E_0). All the laminates showed the same kind of plots, where E_{dis} increases quadratically with the impact energy. The penetration impact energy of each laminate was extrapolated by forecasting the point of intersection between quadratic curves fitted to the dissipated energy discrete values for each laminate and the incident energy 1:1 line [10].

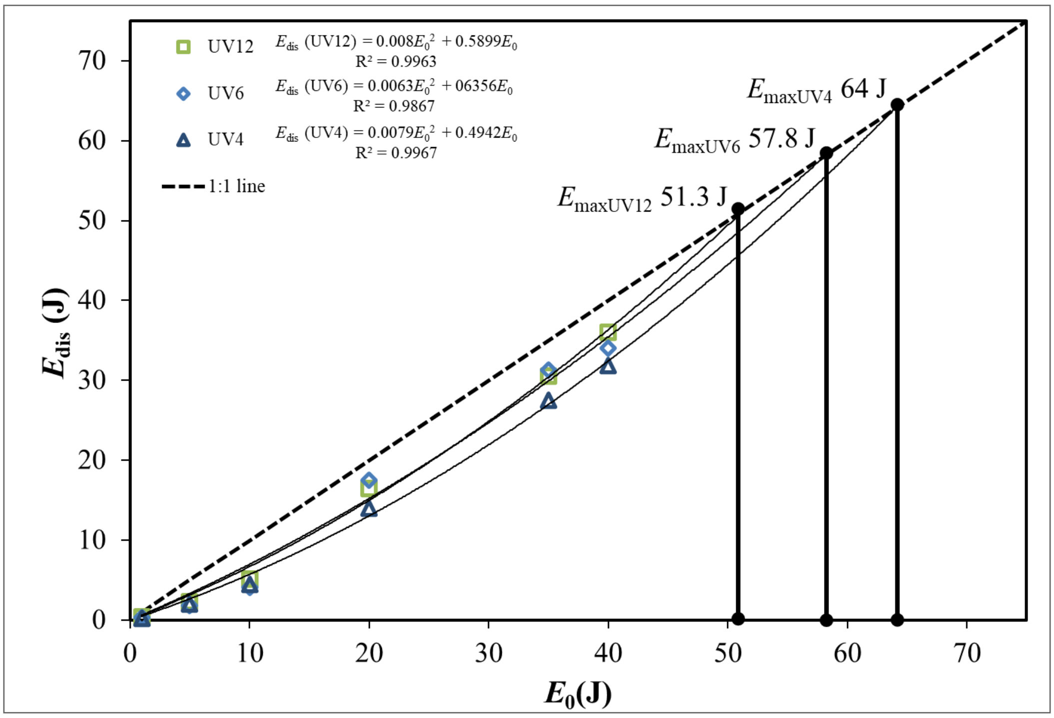

Figure 2. Dissipated energy (E_{dis}) as a function of impact energy (E_0).

As can be appreciated, the UV4 laminate shows the highest penetration energy threshold, and the UV6 laminate improves the results achieved by the laminate UV cured in one shot. This outcome could be related to the fact that these two laminates present bonds that may be more prom to develop delaminations, dissipating more impact energy before initiating other damage micromechanisms. However, if that was the case, the lower interlaminar toughness of the bonds would have negatively affected the bending strength of the UV4 and the UV6 laminates, but they showed better flexural properties. Therefore, the better impact performance of the UV4 and UV6 laminates has to be related to the fact that these layer by layer UV curing strategies induce less residual stress. Furthermore, it can be concluded that the thinner the thickness cured in each UV exposure is, the lower the generated residual stress would be.

4. Conclusions

The feasibility of the layer by layer *in-situ* UV curing processing of glass fibre reinforced polyester composites has been demonstrated in the present paper. The flexural stiffness and strength of the composites UV cured following a two step and a three step strategy resulted to be higher than for the laminate consolidated in one shot. Additionally, the impact energy dissipated in the low velocity impact tests was higher the thinner the layers cured in each step had been, so the best performing laminate was that cured in three steps, followed by the one manufactured in two steps, and the laminate cured in bulk presented the worse results.

Thus, two conclusions that could help selecting the most appropriate layer by layer manufacturing strategy can be extracted from this study. On the one hand, that the intralayer bonds in the layer by layer UV cured laminates are tough. And on the other, that layer by layer UV curing processes induce less residual stress to the composite the thinner the applied layers are.

Acknowledgments

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References

[1] I. Tena, I. Sáenz-Domínguez, M. Sarrionandia, and J. Aurrekoetxea, “Comparación entre el curado ultravioleta y el infrarrojo,” Universidad Rey Juan Carlos, Servicio de Publicaciones, 2015.

[2] N. Li, Y. Li, X. Hao, and J. Gao, “A comparative experiment for the analysis of microwave and thermal process induced strains of carbon fiber/bismaleimide composite materials,” *Compos Sci Technol*, vol. 106, pp. 15–19, Jan. 2015, doi: 10.1016/J.COMPSCITECH.2014.10.008.

[3] A. Endruweit, M. S. Johnson, and A. C. Long, “Curing of composite components by ultraviolet radiation: A review,” *Polym Compos*, vol. 27, no. 2, pp. 119–128, Apr. 2006, doi: 10.1002/PC.20166.

[4] P. Compston, J. Schiemer, and A. Cvetanovska, “Mechanical properties and styrene emission levels of a UV-cured glass-fibre/vinylester composite,” *Compos Struct*, vol. 86, no. 1–3, pp. 22–26, Nov. 2008, doi: 10.1016/J.COMPSTRUCT.2008.03.012.

[5] J. M. Park, J. W. Kong, D. S. Kim, and J. R. Lee, “Non-destructive damage sensing and cure monitoring of carbon fiber/epoxyacrylate composites with UV and thermal curing using electro-micromechanical techniques,” *Compos Sci Technol*, vol. 64, no. 16, pp. 2565–2575, Dec. 2004, doi:

10.1016/J.COMPSCITECH.2004.05.017.

[6] W. Shi and B. Rånby, "UV curing of composites based on modified unsaturated polyester," *J Appl Polym Sci*, vol. 51, no. 6, pp. 1129–1139, Feb. 1994, doi: 10.1002/APP.1994.070510619.

[7] A. Endruweit, W. Ruijter, M. S. Johnson, and A. C. Long, "Transmission of ultraviolet light through reinforcement fabrics and its effect on ultraviolet curing of composite laminates," *Polym Compos*, vol. 29, no. 7, pp. 818–829, Jul. 2008, doi: 10.1002/PC.20483.

[8] A. Di Pietro and P. Compston, "Resin hardness and interlaminar shear strength of a glass-fibre/vinylester composite cured with high intensity ultraviolet (UV) light," *J Mater Sci*, vol. 44, no. 15, pp. 4188–4190, Aug. 2009, doi: 10.1007/S10853-009-3544-2/FIGURES/2.

[9] I. Tena, J. A. Arakama, M. Sarrionandia, J. Aurrekoetxea, and J. Torre, "Effect of thickness on the interfacial strenght of layer by layer in situ UV curing," in *ECCM15-15 TH EUROPEAN CONFERENCE ON COMPOSITE MATERIALS*, 2012.

[10] P. Feraboli and K. T. Kedward, "A new composite structure impact performance assessment program," *Compos Sci Technol*, vol. 66, no. 10, pp. 1336–1347, Aug. 2006, doi: 10.1016/j.compscitech.2005.09.009.