On the influence of polymer surface layer thickness on the adhesion of composite assembly. Differences between initial state and thermal ageing.

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Introduction

Bonding of composite materials with an adhesive layer is one of the most promising alternatives to classical bonding techniques like bolting or welding. In order to increase the performance of composite assemblies several surface treatments have been developed [1-2]. Then some authors previously demonstrated the effectiveness of reducing the thickness of the polymer surface layer [3-5] on adhesion performances. But conclusions of these results at the initial state would not be obviously applicable to several ageing conditions.

Experimental

Composite materials. Carbon/epoxy and glass/epoxy composites were manufactured by AIRCELLE Le Havre (SAFRAN Group, FRANCE) using an autoclave process. Carbon/epoxy samples are made of 10 superimposed plies, in the same way glass/epoxy samples are realized also with 10 carbon/epoxy plies with one more glass/epoxy ply on the bonded surface.

Surface treatments. In order to increase or reduce the thickness of polymer surface layer, two surface treatments are compared to the peel ply surface treatment which can be considered as reference sample. The first surface treatment that reduces the polymer thickness is an excimer laser ablation that allows, in specific conditions, an ablation of epoxy matrix without any degradation of fibre reinforcement. The second surface treatment is a tear ply treatment that creates a rough and clean surface, the main difference with a classical peel ply treatment is that tear ply is previously impregnated with polymer matrix. This impregnation allows an increase of polymer surface thickness.

Single lap shear tests. Single lap shear tests are performed on 100mm×25mm specimens with a 12.5mm×25mm bonded area. Two different epoxy adhesives (A1 and A2), commonly used industrially, are chosen to realize bonded assemblies. Shear stress measurements are carried out at room temperature at 2 $mm.min^{-1}$ constant displacement rate on an *INSTRON 4204*. The maximum stress necessary to break the joint is determined from the stress/time curves, on five assemblies.

Light and electron microscopy. This study is performed with *LEICA DM LP* which enables observations from 50× to 500× with polarized or non polarized light. And on the other hand a scanning electron microscope *HITACHI S 3000 N* within magnitude from 30× to 2500× is used.

Thermal ageing. The thermal ageing is performed in a *Kendro Heraus* (model Kelvitron UT 6120). This apparatus allows the temperature to be maintained at 150°C with homogeneity of 3°C (with the air trap shut).

Results and discussion

Initial state. Presenting authors previously demonstrated that when a strong bond is established between an adhesive layer and a composite surface, the failure mode is mainly due to a cohesive failure inside the composite material between fibre reinforcement and the polymer surface layer [3-4]. Indeed in this case there is no need to use several surface treatments as the material itself becomes the weak point of the bonded assembly. Hence, peel ply and tear ply treatment exhibit very similar lap shear values owing to their cohesive failure inside the material.

One possible alternative is then the use of ablation treatments in order to bond directly the fibre reinforcement by reducing the size of polymer matrix layer. Then several ablation modes are used according to an excimer laser surface treatment. Results show that a partial ablation of the polymer surface layer can increase very significantly adhesion performances, with a modification of failure mode from cohesive in the composite to cohesive in the adhesive. But this result at this initial state is not obviously the case after different ageing conditions.

Thermal ageing. Indeed previous works have shown that the degradation occurring during thermal ageing mainly comes from micro/macro voids coming from the adhesive layer [6]. Hence the degradation from these voids to the whole adhesive layer is mainly governed by several parameters such as fillers, supported films… Then it appears from transversal micro cutting that the degradation shifts in an easier way from the adhesive to the composite material if there's locally a richer area in matrix on the composite surface. Indeed, fillers from the adhesive or fibre reinforcement from the composite may contain the degradation inside the adhesive. Though the degradation develops more easily in polymer matrix surface layer.

This hypothesis is then confirmed with single lap shear tests and microscopic observations of tear ply sample compared to peel ply one after 1000 hours ageing at 180°C. Indeed, the degradation of the adhesive is significantly lower for tear ply samples, this is probably due to an easier shifting of the degradation from the adhesive to the composite material which owns a high thickness of polymer on its surface.

Conclusions

The influence of polymer surface layer is of primary interest as it can completely govern the adhesion behaviour of composite assemblies at the initial state and after several ageing conditions.

Then the interest of reducing the thickness of polymer surface layer is shown as it can completely increase the adhesion performance of carbon/epoxy and glass/epoxy samples. Indeed it allows changing the failure mode from cohesive in the composite to cohesive in the adhesive.

However this conclusion at the initial state can not be linked with the behaviour of composite assembly after thermal ageing. Indeed, as the degradation mainly comes from the micro/macro voids of the adhesive layer, the degradation of both the adhesive and the bonded performances can be limited with the use of higher thickness polymer surface composite. Then tear ply treatment that allows an increase of polymer matrix on the composite surface can avoid the degradation of the adhesive layer by shifting the degradation from the adhesive to the composite material itself.

All these results mainly confirm the influence of micro/macro voids on the degradation of composite assemblies. But it also explains the main influence of fillers, composite surface and adhesive layer on the whole stability of the bonded assembly.

References

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