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Experimental investigation of hydrothermal ageing on single lap bonded CFRP joints

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Abstract

Composite materials are increasingly being used in various field of engineering interest over recent decades. As an alternative to fastening, bonding is one of the most promising assembly techniques of composite materials since it allows a uniform distribution of forces, it has a greater ability to dampen vibration and it does not raise any problems of corrosion typical of metal fasteners.
Currently the use of composite materials is limited by the incomplete knowledge of their behaviour in an aggressive environment. For example, the factors influencing the durability of the bonded joints are mainly temperature and humidity, but it is usually impossible to predict their effect without performing experiments. In this work is investigated how the hydrothermal ageing can affect the mechanical resistance of CFRP single lap joints. The parameters chosen for the activity consist in two types of adhesives (AF 163-2K film and EA 9309NA paste) and three ageing environments (thermal cycles from -28 °C to 85 °C in air, distilled water and salt water).

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Keywords: Hydrothermal ageing; Adhesive bonding; Single lap joint; Experimental tests.

1. Introduction

Compared to traditional fastening systems, the use of structural adhesives allows obtaining a series of advantages such as the significant reduction of the weight of the joints, the elimination of very dangerous corrosive problems in the case of rivets and metal bolts and the reduction of the local delamination of the parts due to drilling, as noted by Sorrentino et al. (2017 and 2018b).

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However, nowadays their use is limited by the incomplete knowledge of how they can behave over time in contact with more or less aggressive environments. For example, it is known that in the case of epoxy adhesives the factors of influence of the stability of the joint over time are mainly the temperature and humidity, but in most cases it is impossible to estimate their effect on the durability of the joint except by experiments. In seawater, the galvanic coupling due to the different nature of the adherends not only causes the onset of corrosive phenomena on the metallic adherence, but can also lead to forms of damage in composite adherend like bubbles and swellings, as noted by Tucker et al. (1989). Both in steam and in liquid form, water is one of the most aggressive environments for bonded joints. This peculiarity generally derives from the presence of polar groups in the adhesives, which make the bonding intrinsically hydrophilic. Bowditch (1996) has observed that the addition of silicon in the adhesive is able to decrease the amount of mechanical degradation due to immersion in water for a certain period: the adhesive becomes more stable in water. He also states that increasing the test temperature to accelerate itself can be misleading because it is easy to analyze different damage mechanisms in this way. In fact, often the aggressiveness of the test environment is increased (for example by raising the temperature, increasing the humidity or simply varying the chemical composition), in order to reduce the test time and often neglecting the true phenomena that would occur in the real case. Armstrong (1996 and 1997) has studied the durability in distilled water of bonded joints realised with aluminium adherends combined with various types of epoxy adhesives. Specifically, he observed that the degradation is directly proportional to the diffusibility and to the solubility of the water in the adhesive. Wylde and Spelt (1998) have analysed the effects of temperature on water diffusion in epoxy adhesives, observing that the saturation condition is a function of temperature as long as it is below the T_g. Obviously the different nature of the adhesives does not make it possible to generalize the observations made only on specific tests. Wilken et al. (2005) have experimentally observed that the presence of an interface between adherend and adhesive amplifies the diffusive phenomenon. In their work, Zhou et al. (1995) have observed that a composite laminate immersed in water for a sufficiently long time may show a partial dissolution of the matrix, observable only by gravimetric analysis. Still in the same work, Zhou et al. have noted that the expansion due to the absorption of water is much more limited in the direction of the fibres respect the others. Zanni et al. (1995) have analysed the diffusion of water in epoxy adhesives. They noted that the diffusivity is related to the temperature according to Arrhenius law, and that the elastic modulus of the adhesive decreases as the diffusion phenomenon progresses. More recently, McConnell et al. (2010) carried out dielectric studies on the effects of freezing and hydrolytic ageing on bonded joints in composite material realised with this type of adhesive. Using gravimetric analysis, they observed that the composite adherends have a water diffusion coefficient that is one order of magnitude higher respect the adhesive and that the freezing causes an increase in voids and micro-cracks in the adhesive itself.

The research presented in this work is part of an experimental campaign on composite-composite single lap joints. In the previous work of the authors (Sorrentino et al., 2018a), the influence of the surface treatment of the adhesives on the mechanical strength of the joint was analysed. As reported in the literature, the effect of humidity or temperature on bonded joints is known, but the results obtained are not always clear if these two factors act simultaneously. The study of the combined effect of hydrothermal ageing and operating temperature on the strength of the bonded joint is a topic that still needs further investigation. The objective of this second experimental phase is essentially to evaluate how the bonded joint, made of CFRP and epoxy adhesives, reacts to an ageing induced by hydrothermal stress. To obtain a uniform and repeatable bonding surface, all the adherends have been realised with peel ply treatment.

2. Materials and Methods

According to ASTM D5868 and ASTM D1002, the specimens were obtained from two laminates measuring 25,4mm x 101,6mm x 2,54mm while, in order to have a square bonding area, the overlap length was placed at 25,4mm.

The nominal thickness of the adhesive was set equal to 0,76mm. A prepreg produced by SAATI S.p.A., made of ER450 epoxy resin and CC289 carbon fibres with 5H satin weave, was used for the realization of the adherends. The layup used was $[(0°, 90°)]_8$, where (0°, 90°) is one layer of fabric. The treatment process consists of a ramp of 2 ° C / min. for 55min. and in a maintenance at 135 ° C for 2 hours at a pressure of seven bar. The peel ply used is 51789 produced by Precision Fabric Group, made of nylon fibres woven together. The operations carried out to obtain the surface finish simply consist of inserting it at the end of stratification on the laminate and removing it at the end of the polymerization just before the bonding phase.

The adhesives identified to perform the tests are the EA 9309NA paste adhesive produced by Hysol and the AF 163-2K film adhesive produced by 3M Scotch-Weld. The Hysol EA 9309NA is a two-component paste adhesive, composed of a translucent beige paste (part A) and a red activator (part B), to be mixed according to the weight ratio of respectively 100 (part A) and 23 (part B). For this adhesive, the polymerization reaction takes place at room temperature for a period of 7 days. The AF 163-2K is a film adhesive with a thickness of about 0,24mm and has a carrier inside which gives it greater tensile strength. The polymerization reaction occurs in an autoclave according to the technical data sheet. In total, 18 specimens were made, which were divided into 6 lots respectively, each consisting of 3 specimens. The 6 lots were randomly divided into three groups, so that each group has the same number of joints made with AF 163-2K and EA 9309NA. Subsequently, the created groups, made up of six specimens each, were subject to different cyclical conditions of ageing (Table 1).

Table 1: Experimental Plan

Factors	# Level	Levels
Adhesive	2	AF 163-2K, EA 9309NA
Ageing conditions	3	Distilled water, salt water, air
Replications	3	

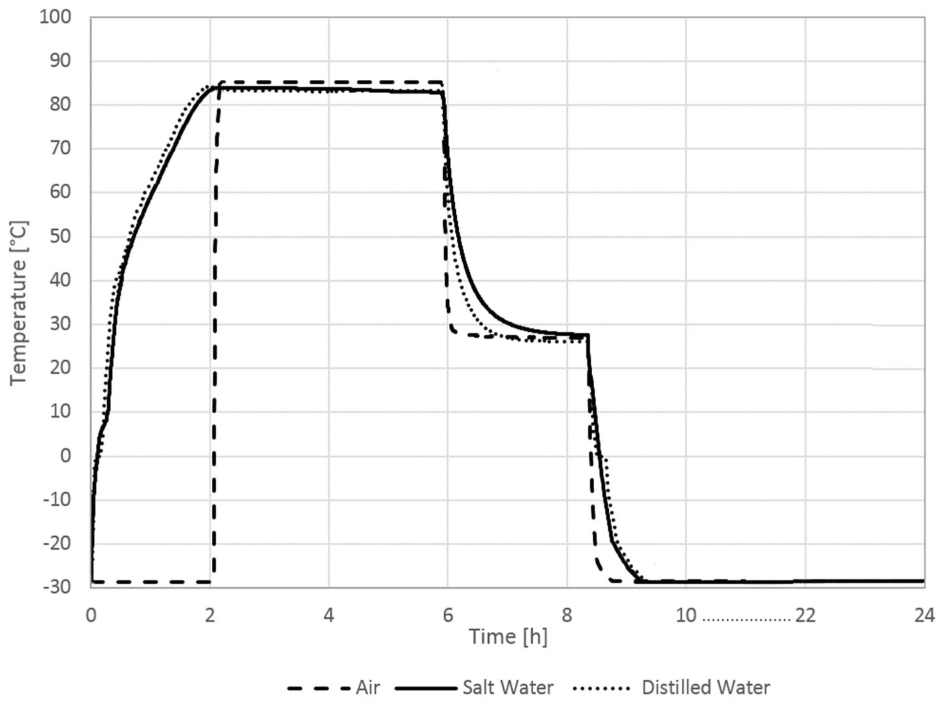

Fig. 1. Ageing cycles measured with K-thermocouple.

To realise the ageing treatments, it has been used a model 125 / 45HA oven manufactured by Nabertherm GmbH, equipped with a convection chamber; and a 2700 model multimeter manufactured by Keithley Instruments, combined with the use of K-type thermocouples for temperature control during the process;

The specimens relative to the first group were subjected to a daily cycle of 4 hours at 85 ° C and about 20 hours at -28 ° C. The specimens of the second group were immersed in distilled water. In order to avoid evaporation of the water, which would cause the rise of the specimens from the water itself, the container was closed with a nylon film called elastomax produced by Aerovac Systems Ltd. The specimens relating to the third group underwent the same process applied for the second group, using salt water instead of distilled water. The saline solution was made according to ASTM D 1183. The latter groups were subjected to a daily freezing cycle at -28 ° C for 16 h and thawing and maintaining at 85 ° C for 6h. The remaining 2 hours were used to cool the containers to an acceptable temperature in order to place them in the climatic chamber (Fig. 1). At the end of each cycle, a check on the water level in the containers was performed and, where necessary, a top-up was carried out.

In order to monitor the temperature, a laminate of the same thickness of the adherends was inserted into each group with a K-type thermocouple inside. All three groups have undergone the above-mentioned cycles for thirty days.

Fig. 1 shows the trend of the temperatures in the respective environment of the three groups. The heating transient for the two groups in water was completed within two hours. Subsequently, the group aged in air was placed in the oven and, having a much lower thermal inertia, reaches the operating temperature within a few minutes. Even in the cooling phase, the different thermal inertia between the three groups involves different cooling speeds: specifically, the cooling rate of the samples in air was an order of magnitude higher than the cooling speed of the samples in water.

Once the ageing phase was completed, the specimens were removed from the containers and left to dry at room temperature for one week, then they were tested.

The test conditions were performed according to ASTM 5868-01 using an INSTRON 5586 with a maximum capacity of 300kN and setting a crosshead speed of 1.3 mm/min.

3. Results and Discussions

In order to have a reference to the failure load of the non-degraded specimens for both adhesives, the results obtained in Sorrentino et al. (2018a) are considered. The results obtained from the lap shear tests are reported in Fig. 2.

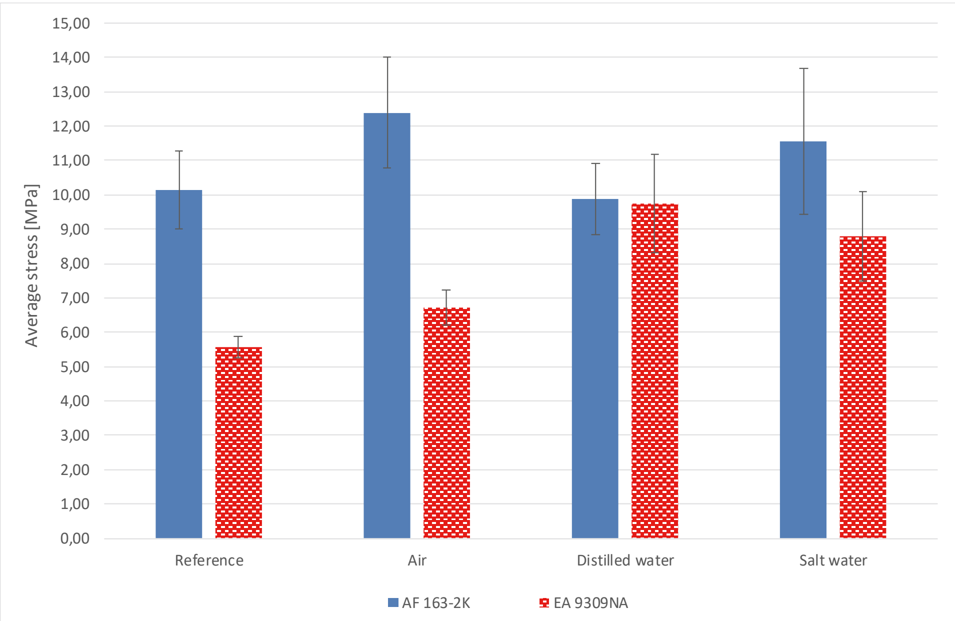

Fig. 2: Results of the tensile tests of the single lap joints.

By analysing the data of the joints made with the AF 163-2K adhesive, it is possible to state that the resistance values are very close to the reference value regardless of ageing, therefore the ageing cycles did not produce deleterious effects for this type of adhesive. The failure modes that show these specimens are all of the Light-Fibre-Tear type, according to ASTM D5573, as shown in Fig. 3.

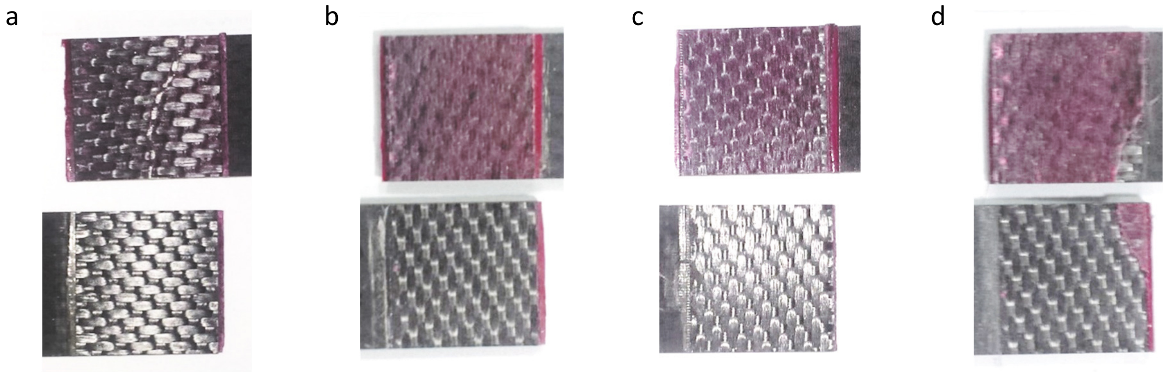

Fig. 3: Failure surfaces of AF 163-2K specimens: a) without ageing; b) with air ageing; c) with distilled water ageing; d) with salt water ageing.

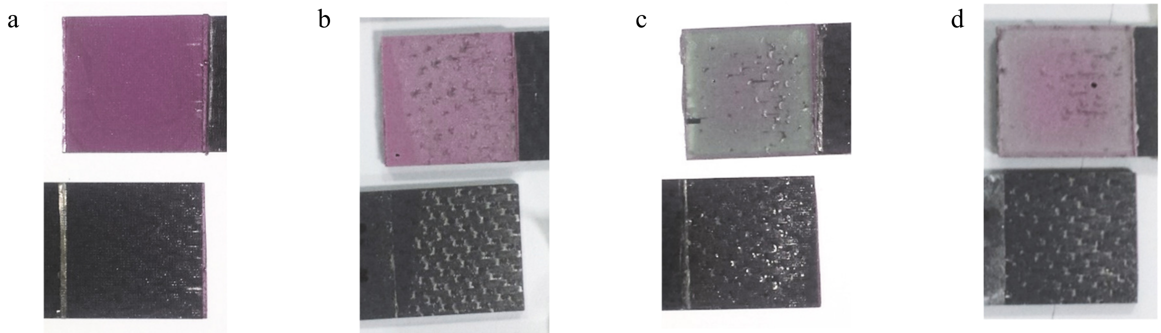

Fig. 4: Failure surfaces of EA 9309NA specimens: a) without ageing; b) with air ageing; c) with distilled water ageing; d) with salt water ageing.

Analysing the data referred to the joints made with the paste adhesive EA 9309NA, it is possible to state that the behaviour of the specimens has been heavily influenced by the ageing conditions, showing variable failure morphologies depending on the type of ageing. Joints aged in water undergo a strong increase in performance, achieving average performances of about 75% higher than the reference ones; also for samples aged in the air there is an increase in performance (due to a post-cure effect induced by the maximum temperature reached during ageing) but less significant (about 35%). Probably in this group of specimens, the effects of damage due to the thermal shock suffered during the transient from - 28 ° C to 85 ° C of the duration of few minutes have taken place, while the specimens aged in water have undergone a softer transient lasting about 2 hours. Furthermore, this difference between the two groups can be due to a greater ductility and lower mechanical resistance of the bonded edges due to degradation in the water-aged specimens, which may have resulted in an increase of the apparent resistance of the aged joints than the non-degraded joints. Chadegani et al. (2011) and Kumar et al. (2013) state that the stress field that is generated inside a single-lap joint has a singularity to the adhesive/adherend interface near the extreme edges, so that the ends of the bonding tend to be stressed more respect the central area of the interface. A greater ductility of the adhesive localized to the extremes allows to attenuate the presence of this singularity and in this way generate a more uniform load, allowing an increase in the apparent resistance of the joint, as noted by da Silva et al. (2009). It is possible to observe the chromatic variation on green that occurs in the joints aged in water (Fig. 4c and Fig. 4d) compared to the other joints tested (Fig. 4a and Fig. 4b), which tends to concentrate near the edges of the bonding area. From the analysis of the failure surfaces, it is possible to observe how the failure mode passes from adhesive (Fig. 4a) to mixed adhesive / light fibre tear (Fig. 4b, Fig. 4c and Fig. 4d): this is probably due to an improvement of the resistance of the adhesive/adherend interface due to ageing.

The results of lap shear test obtained with EA 9309NA have shown a strong influence from ageing. DSC analyses were carried out to analyse how much the ageing has influenced the properties of the adhesive. The heating ramp used is 10 ° C / min. and the observation temperature range is from 25 ° C to 145 ° C. From the DSC results, it is possible to state that the polymerization at room temperature does not allow the total cure of the adhesive. In fact, it present an endothermic peak probably due to some unreacted components. In particular, there are no sensible difference between the behaviour of the edge and the bulk of the bonded area of the adhesive. By analysing the DSC results obtained on specimens aged in air, the adhesive no longer present an endothermic peak. This probably occurs because this treatment has entailed a complete polymerization of the adhesive: in fact, the lack of peaks is compatible with a post cure effect of the ageing. The DSC analysis on the samples tested in water showed that there are different behaviour between the edge and the bulk of the adhesive. In particular, the edge of the adhesive presents an endothermic peak similar to unaged specimens, while the bulk of the adhesive does not present peaks. The adhesive has absorbed water only in the edges and probably this did not allow cross-linking of unreacted components during maintenance at the higher temperatures of the ageing cycle. This phenomenon appears only on the edge of the adhesive because the duration of the ageing treatment was too short to allow a uniform diffusion of water in the entire adhesive. The specimens aged in salt water showed an intermediate behaviour between those aged in distilled water and those aged in the air. This result may be due to a greater difficulty of salt water to penetrate into the adhesive.

4. Conclusions

In this work single lap bonded joints in composite material were made with two types of adhesive: film (AF 163-2K) and paste (EA 9309NA). The specimens were subjected to ageing cycles in air, distilled water and salt water with temperatures between -28 ° C and 85 ° C, and then tested. Regarding the joints made with the AF 163-2K, ageing did not particularly affect the apparent shear strength. With regards to the specimens made with the EA 9309NA, ageing has played a greater role in the apparent shear strength of the joints. In particular an increase in the apparent resistance of the joint aged in air was observed, due in part to a post-cure effect of ageing on the adhesive; while in the case of joints aged in water the apparent resistance is directly proportional to the degradation of the adhesive. This has occurred because the degradation is more localized in the edges of the bonding area, which is where this type of specimen has a stress singularity: in this way, the adhesive has a lower resistance but a greater ductility where such properties are required.

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