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Environmental and chemical degradation of carbon/epoxy lap joints for aerospace applications, and effects on their mechanical performance

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ABSTRACT

Composite structures in service are typically subjected to different environmental conditions, which may affect their mechanical performance as well as their flammability potential. This study investigates the degradation encountered by lap joints made with woven carbon/epoxy and epoxy-based adhesive, treated in water or anti-icing additive or jet fuel or hydraulic fluid. Gravimetric tests were carried out, and degradation was observed with changes in color, mass, microscopic features and mechanical properties, with the support of statistics. This investigation proves that the performance of the epoxy-based structural adhesive deteriorates considerably when the adhesive is contaminated by hydraulic fluid and anti-icing additive, while the carbon/epoxy weave appears affected by anti-icing additive at a lesser rate.

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1. Introduction

Composites have been used since the 1970s for applications requiring high stiffness/weight and strength/weight. They have been gaining much attention recently because of their use in load-carrying parts of new aircraft such as the Boeing 787, Airbus 350 and F-35. They are also becoming more common in non aerospace applications, for example in ships [1], in retrofitting of structurally-deficient bridges, in the construction of new pedestrian and vehicular bridges [2], and for transportation (e.g. cars [3], and trains [4,5]).

Composite parts can be assembled with fasteners, adhesives or a combination of both. The advantages of bonded joints include: (1) reduction of local delamination because no holes are required, (2) significant reduction in the weight of the joints, the ability to, (3) assemble dissimilar materials, (4) prevent galvanic corrosion of conductor metals, (5) design a smooth external surface. Bonding of parts is required not only during manufacture but also during composite repairs, where damaged skin is removed and replaced by new layers of epoxy-impregnated fabric.

This study focuses on the environmental and chemical degradation of the components of lap joints. Lap joints are commonly reported in the literature and in the engineering practice as an easy-to-model example of bonded joint, e.g. [6,7]. It was shown as early as 1959 that the shear stress in adhesives is increased by solvents [8]. Many researchers have characterized the degradation of carbon- or glass-reinforced polymeric composites by exposure to different environments: water, saline water, acidic water, organic fuel, low and high temperatures (to name a few [9–15], and review papers [16,17]). Polymer degradation also affects the flammability properties and rises the risk of pyrolysis (e.g. [18], and the follow-up paper written with collaborators from the Fire Research Team of the US Air Force Research Laboratory [19]).

Disbonding due to hydraulic fluid was found on commercial aircraft, in particular the composite rudders of some Airbus A300 series aircraft. The US National Transportation Safety Board reports in 2006 that '*Contamination with hydraulic fluid will lead to a reduction in the bond strength and an overall loss in the rudder's structural integrity*, [...] *and leaves the airplane susceptible*' to a type of in-flight rudder separation [20]. In addition, it was suggested in [21] that the design of composite bonded repairs should also take into account the presence of anti-icing additive.

Adhesive manufacturers publish limited data on their products' performance due to environmental/chemical effects. For example, the technical data of a structural adhesive from 3 M shows reduced overlap shear strength after exposure to air, anti-icing fuel additive, hydraulic fluid and jet fuel, for up to 90 days [22]. The study discussed in this paper will show that a 90 days period may not be sufficient for an accurate assessment of strength. Moreover, this assessment may be further complicated by high temperature fluctuations typical of operational aerospace missions, e.g. −55 °C and runway temperature for subsonic flights, and −55 °C and 130 °C for supersonic flights [23]. This aspect, as well as the potential compounded effects of thermo-mechanical fatigue and chemical/environmental degradation, will not be addressed in this paper.

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Moreover, commercial sizing may mitigate or complicate matters, e.g. [24], but it has not been considered by the authors of this paper.

Unmanned aerial vehicles, UAVs, provide a low-cost example of the potential dangers of water and artificial fluids on composites. Fig. 1 shows aircraft damage due to jet fuel and water in a UAV. These locations cannot be easily inspected unless technicians remove the appropriate panels. This emphasizes the difficulty of detecting aircraft problems such as these by visual inspection, the most common type of nondestructive inspection in aviation.

Even though much research has been carried out to date on the topic, the physical and chemical mechanisms affecting the durability of composites are complex, and concern fibers, matrix and fiber/matrix interphase (as defined in [17]), at different rates and with different triggering temperatures and reactions. An adequate and practical solution to identify, quantify and repair the damage due to environmental conditions and exposure to chemicals has not yet been found.

The results presented in this paper appear novel and provide further insight into the problem. In particular, this investigation documents the deleterious effect on strength of structural adhesive due to hydraulic fluid and anti-icing additive. On the other hand, carbon/epoxy specimens appear damaged especially by anti-icing additive. Jet fuel and water are much less detrimental, by comparison. This paper has the potential of improving safety and design requirements in all engineering applications adopting load-carrying composite structures.

2. Manufacturing and testing processes

To understand how each component of a bonded joint is affected by contaminants, gravimetric experiments were carried out on lap joints made with woven carbon/epoxy and aerospace-grade adhesive, followed by tests on adhesive specimens and monolithic carbon/epoxy.

2.1. Single lap joints: manufacture, gravimetric and tensile tests

A preliminary study was carried out on single lap joint made with woven carbon/epoxy adherends [25]. Composite panels were manufactured with ten layers of T-300 plain weave with weight/area equal to 98.3 g/m^2, producing a thickness consistent with ASTM D5868-01 [26]. The panels were infused with low-viscosity Proset® 117 LV epoxy mixed with Proset® 237 hardener [27], using a conventional, low-cost out-of-autoclave Vacuum Assisted Resin Transfer Molding, with a cure cycle of 4 h at 50 °C followed by 16 h at 60 °C. The resin/hardener mix was degassed before the cure, to minimize the formation of bubbles. The measured material properties of the weave turned out to be much lower than those published by the manufacturer, so this material was discontinued after this preliminary study [25].

The lap joints were manufactured with a 25.4 mm overlay area (as recommended by ASTM D5868-01 [26]) in a 120 mm gauge length specimen. The adhesive chosen for the study, Hysol® EA 9360, [28], is utilized in aerospace applications, and has high peel strength and a nominal service temperature of 107 °C. The service temperature is defined by the manufacturer to be the temperature at which an expected strength percentage is retained. The adhesive was cured for 5 days at room temperature, consistently with the technical data sheet. Tabs were cut out of G10 fiberglass and applied on the specimens for subsequent tensile tests. Then, the specimens were fully soaked (100% relative humidity, RH) at room temperature for 243 h, in: (a) fresh water, or (b) jet fuel, or (c) Skydrol® 500B hydraulic fluid, or (d) Prist® Hi-Flash™ anti-icing fuel additive. The anti-icing aviation fuel additive, diethylene glycol monomethyl ether, controls icing in aircraft fuel by lowering the freezing point of the water present in the fuel. Usually it is mixed with jet fuel, but in this experiment 100% fuel additive was used to simulate the worst case scenario. Five specimens were prepared for each test condition, in addition to six baseline specimens. No sealant was used for the specimens' edges. The time of 243 h was due to a time constraint. While this time is short, it allowed the authors to identify questions, challenges and improved procedures needed for further iterations of this study, as shown in this paper. Moreover, the time is consistent with some data available by the adhesive manufacturers (e.g. 3 M reports data of adhesive shear strength after only 7 days of immersion in anti-icing and hydrocarbon fluid [22]). The ASTM D5229 standard [29], was adopted for the gravimetric tests, as it addresses moisture absorption/desorption, with moisture being intended as '*liquid (water, jet fuel, salt water or any other liquid) which is* [...] *present in quantity sufficient for immersion of an object*'. The mass gain was calculated from

$$M(\%) = \frac{\text{mass of wet specimen} - \text{mass of dry specimen}}{\text{mass of dry specimen}} \times 100.$$

A Mettler balance with a 100 g range and resolution of 0.1 mg was utilized. The specimens were tested under uni-axial tension with an Instron 4204 screw-driven machine, at a rate of 1.3 mm/min. Fig. 2 summarizes key results of this preliminary investigation. The mass gain plot shows that the test time was clearly not sufficient for equilibrium of the specimens immersed in anti-icing fluid. Even within the given time frame, anti-icing additive was absorbed from the lap joint at much higher rates than the other fluids. A two-tailed *t*-test with unequal variance and 95% confidence was applied upon verifying that the data was normally distributed (through Shapiro–Wilk test for normality). The *t*-test showed that all specimens (treated and control) belonged to the same distribution. The result is confirmed by the box plot in Fig. 2.

The main conclusions that can be drawn by this preliminary work are the following:

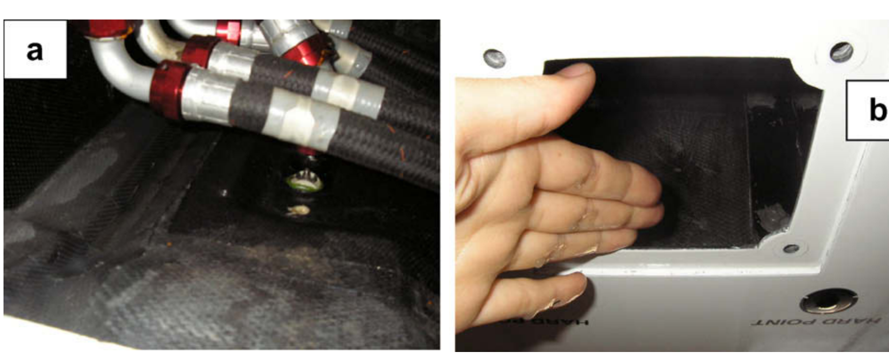

Fig. 1. (a) Fuel leak from fuel tank/hoses, in UAV, and (b) water condensation inside UAV wing.