Effect of Corrosion on Mechanical Properties of Adhesive Joints

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Abstract

Compared with conventional mechanical joining techniques, the use of adhesive bonds presents numerous advantages such as a more homogeneous stress distribution, higher stiffness, high fatigue strength, low weight, the possibility to join dissimilar materials or corrosion prevention for these reasons, structural adhesive bonding has been increasingly used in industries including construction, automotive and sports The identification of the harmful factors and contaminants affecting. The performance of composite joints for aeronautics applications has not been fully achieved. Furthermore, their presence cannot be easily detected by conventional non-destructive tests (NDT). Water diffusion into composite adhesive joints was experimentally studied by means of gravimetric measurements in bond specimens. The main parameters controlling the diffusion in the bond components were obtained from water uptake experiments carried out in bulk specimens of fibre-reinforced composite and epoxy adhesive. The diffusion of water in bonds is a complex mechanism due to the heterogeneous microstructure of the composite and its interface with the adhesive. This process was simulated through micro scale models which represent the microstructure of the composite and of the adhesive joint Structural adhesive joints are adequate for joining aluminium with nearly constant. In terms of fatigue strength, for105 cycles, a decrease about 25% and 39% occurred in specimens immersed during.

Keywords: Structural adhesives; degradation of adhesives; water; motor oil.

1. Introduction

Adhesive joints offer advantages relatively to conventional joining processes, namely acoustic isolation, vibration attenuation, reduction of corrosion problems, and a more uniform stress distribution. Also adhesive bonding is a cheap, fast and robust joining technique increasingly used in structural applications, namely in automotive, aeronautic, aerospace, electronics and electric industries [1, 2]. In fact, this technique has obvious advantages; however, its limited ability to withstand the aggressive environments is a considerable restriction for many applications.

The main environmental factors in climatic exposure are temperature and humidity [3]. In terms of temperature, according to Banea *et al.* [4], the most significant factors that determine the strength of an adhesive joint are: The cure shrinkage, the co-efficient of thermal expansion of adhesive and the change in adhesive mechanical properties with temperature. As a consequence of the polymeric nature of the adhesives, the glass transition temperature (T_g) is a very important parameter because T_g establishes the service environment adequate for the materials' usage. At high-temperatures, for example, the load transmission capability of the adhesive joints decreases because the stiffness and strength of the adhesive decreases [5]. Additionally, adhesives suitable for high-temperatures are generally brittle at low-temperatures, giving low joint strengths at low-temperatures, while adhesives suitable for low-temperature are too weak or degrade at high temperatures [6]. In fact, the open literature presents several studies about the effects of moisture and temperature on adhesive joints strength but there are very few works about the effect of highly corrosive environments. Prolongo

and Ureña [7], for example, studied the durability of epoxy-aluminium joints, with a homopolymerised epoxy resin, under a saline environment and observed that the degradation of the joint occurred on the adherends by corrosion. On the other hand, for saline environments, Del Real . [8] showed that the durability of adhesive joints can be increased significantly with surface treatments. Therefore, this work intends to contribute for a better understanding of the effect of corrosive environments on the mechanical properties of single-lap adhesive joints. A saline environment was considered and the adhesive joints performance was analysed by tensile and fatigue tests. Markatos *et al.* identified significant degradations in the fracture toughness of bonded joints in five different harmful scenarios: moisture, release agent and Skydrol contamination, effect of the curing process and high temperature during service. Moisture contamination is probably the most studied scenario for bulk polymers and composite materials [3–6], and numerous models have been proposed to adequately describe this process.

The ASTM standard for water diffusion in thin plates of fiber reinforced polymers (FRP) [9] considers that this process can be adequately described by the one dimensional Fick's second law, which is given by:

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial X^2} \qquad (1.1)$$

where C is the moisture concentration, D is the diffusion coefficient, t is the time and X is the position through the thickness. A useful closed-form approximate solution is given as [10]:

$$\frac{M(t)}{s} \approx 1 - \exp\left[-7.3\left(\frac{Dt}{h^2}\right)^{0.75}\right] \qquad (1.2)$$

where $M(t)$ is the evolution of the water absorbed, s the water content at saturation and h the thickness of the plate. Fick's law is the simplest model to describe the diffusion of water into a material. However, moisture absorption in epoxy adhesives cannot usually be accurately described with this model Thus, more elaborated diffusion theories are required to reproduce the experimental moisture-uptake curves. The most widely accepted diffusion models to reproduce the water absorbed in polymers can be divided in two groups: multiphase diffusion (Langmuir model) and time-dependent diffusion. The Langmuir diffusion model was developed to deal with two phases of different diffusion

kinetics. This model assumes that water can diffuse into the material, but some water molecules are also trapped inside the epoxy microstructure. This behaviour is described by:

$$\frac{\partial C_f}{\partial t} + \frac{\partial C_b}{\partial t} = D\frac{\partial^2 C_f}{\partial x^2} \qquad (1.3)$$

$$\frac{\partial C_b}{\partial t} = \gamma C_f - \beta C_b$$

where subscripts f and b stand for the free and bounded water phases, respectively. The parameters γ and β control the probability of water in each state. As for the Fickian model, the solution can be approximated by [11]:

$$\frac{M(t)}{s} \approx \frac{\beta}{\gamma + \beta}\exp(-\gamma t)\left\{1 - \exp\left[-7.3\left(\frac{Dt}{h^2}\right)^{0.75}\right]\right\}$$

$$+ \frac{\beta}{\gamma + \beta} \times [\exp(-\beta t) - \exp(-\gamma t)]$$

$$+ [1 - \exp(\beta t)] \qquad (1.4)$$

Time-dependent diffusion models are supported by the experimental data which show that both saturation and diffusivity are specimens clearly stress-dependent in some polymer proposed time-dependent boundary conditions to reproduce the water uptake data in epoxy resins. All the aforementioned models are able to fit the experimental water uptake curves in terms of weight gain. However, La Plante *et al.* used nuclear resonance imaging of deuterated water into a polymer to demonstrate that the moisture distribution through the thickness of the specimen could be more adequately fitted by considering time dependent boundary conditions. For this reason, this later approximation was selected in this work for the numerical simulations of the water absorption in the adhesive specimens early stress-dep Material sand.

2. Experimental Testing Methodology

Docol 1000 high strength steel (SSAB, Borlänge, Sweden) plates with 1 mm thickness was the material used for the a dherends of the single –lap joints studied. Theme chemical properties were obtained from tension static tests, performed according with STM E 8M Standard, and are presented in Table 2.1. More details about this material can be found by Cognard *et al.* [12] for adhesive, Figure 2.1.

Material and environmental conditions as detail in Table 2.1, 2.2 and load displacement curve shown in Figure 2.2.
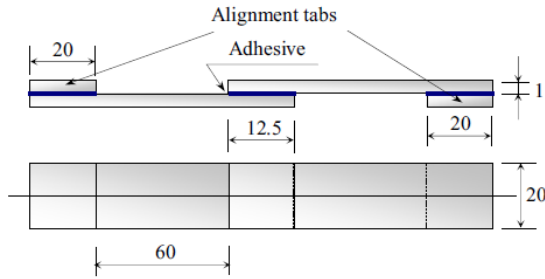

Figure 2.1: Specimen geometry with 150 μm adhesive thickness (dimension in millimeters).
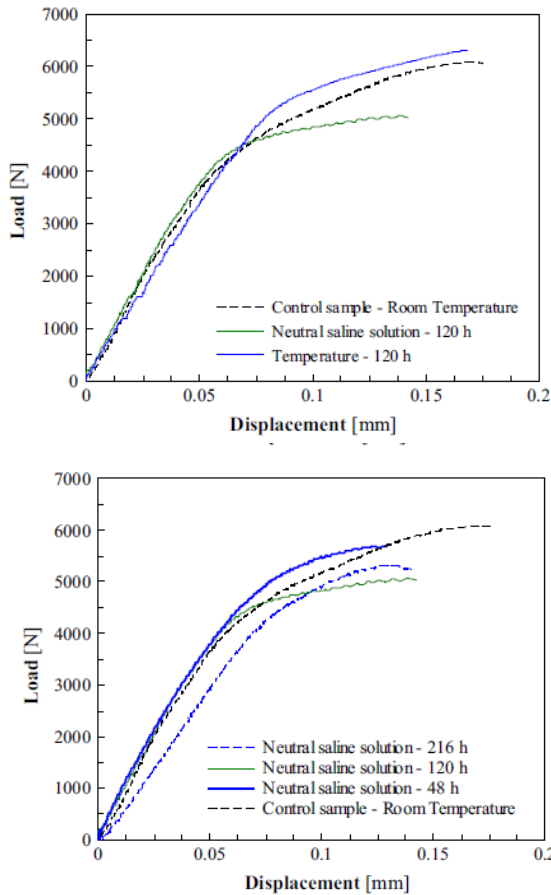

Figure 2.2: Load–displacement curves: (a) joints subjected to different environmental conditions and (b) joints subjected to different time exposures to saline solutions.

Table 2.1: Mechanical properties of the a dherends and adhesive, (σ_{UTS} – tensile strength, σ_{ys} – yield strength, E – Young's modulus, ε_f – elongation at failure, and ν – Poisson's ratio).

	Unit	Material	
		Docol 1000 high strength steel	Adhesive (Araldite s 420 A/B) [12, 13]
σ_{UTS}	MPa	1052.0	35.0
σ_{ys}	MPa	502.0	27.0
E	GPa	205.0	1.85
ε_f	%	11.6	8.5
ν	-	—	0.3

3. Conclusion

The water absorption in adhesive composite bonds was experimentally studied by measuring the mass gained during sample immersion in distilled water at 70 °C. Experimental measurements showed that the diffusivity parameter of the adhesive bond was one order of magnitude lower than the values obtained for the laminate and the adhesive. This effect was a consequence of the reduction in the water flux produced by the presence of the reinforcement fibers. The water absorption experiments were simulated by means of finite elements models which took into account the actual composite microstructure. Simulations predicted the mass gained by the adhesive bond and its components during the water immersion. Numerical results showed the ability of the micro-scale models to capture all the micro mechanisms of water diffusion in composite bonds. Finally, the numerical models were employed to ascertain the influence of the matrix and adhesive diffusivity and the adhesive thickness in the water absorption in adhesive bonds. It was found that the diffusivity of the matrix plays a main role to control the diffusion rate. The thickness of the adhesive has a secondary role, a thicker adhesive layer slightly reduces the diffusion rate and increases the water content in the joint. Thus, the diffusivity of the composite can be considered as an upper limit for the diffusivity of the joint. This approximation is very accurate when the adhesive layer is very thin compared with the composite parts.

This work analyzes the degradation of aluminum-composite adhesive joints under the action of water and motor oil. For this purpose, we have been performed an accelerated aging of the adhesive joint

Table 2.2: Environmental conditions studied.

Series	Environment/exposure	Condition	Exposure time (h)
ND	Control samples	20±2 1C; 50±2% HR	–
WD	Deionised water	35±2 1C; pH:6.7	120
NSS	Neutral saline solution	35±2 1C; pH:7	24/48/96/120/192/216
TEMD	Temperature and relative humidity	35± 1C; 25±2% HR	24/48/96/120/168

by immersion in water and motor oil. Likewise, we have evaluated the loss of mechanical properties that aging causes in the adhesive joint (with ENF tests) Figure 3.1.
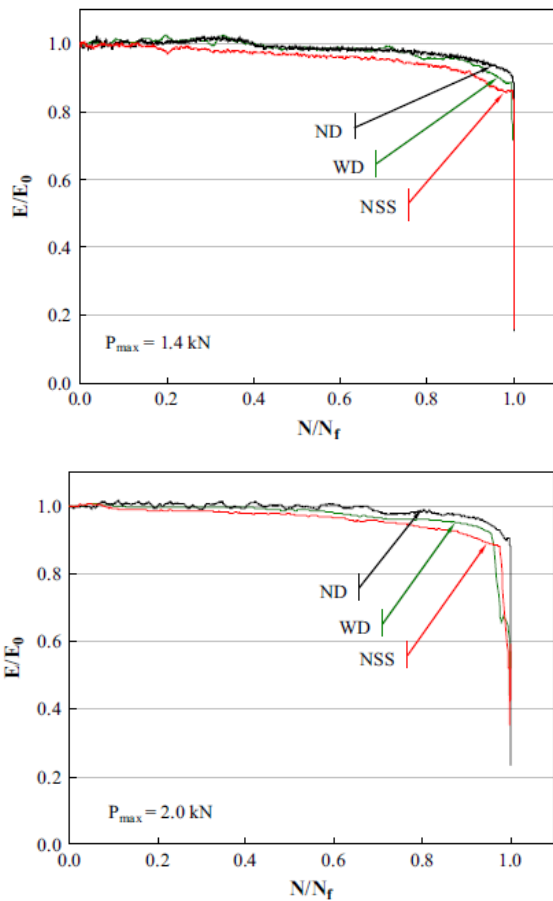

Figure 3.1: E/E0 against the normalized number of cycles N/N_f for: (a) maximum load of 1.4 kN and (b) maximum load of 2.0 kN.

Results of gravimetric tests show that motor oil concentration in the adhesive is lower than water concentration (except during the first days of immersion). Water concentration increases in the adhesive while that oil concentration decreases and stabilizes (0.5%). Additionally, tests have shown that the water spreads more into epoxy that into polyurethane (nearly doubles the concentration of water in the long term). It is also observed that the motor oil concentration into polyurethane during early periods of immersion is lower than into epoxy. However, both concentrations tend to be similar in the long time.

ENF tests (end-notched flexure tests) show the loss of mechanical properties that aging causes in the adhesive joint. This loss of mechanical properties has been evaluated by the percentage reduction of failure load of the adhesive. Considering the relative percentage, the reduction of failure stress in the epoxy is higher than in the polyurethane. With immersion in oil, the reduction is 20% for the polyurethane and 30.7% for the epoxy. With immersion in water, the reduction is 54.4% for epoxy and 23% for polyurethane. The failure stress with immersion in oil is higher than in water (for polyurethane and epoxy). At end of 128 days of immersion and considering relative percentage, the failure of polyurethane adhesive joint with immersion in oil is 16% higher than in water. Similarly, the failure stress of the epoxy adhesive joint immersed in oil is 26.6% higher than in water. In summary, water degrades the adhesive more than motor oil. Additionally and under the action of water or motor oil, polyurethane adhesive joints retain their mechanical properties (failure load) better than epoxy adhesive joints. The tensile static strength and fatigue of single-lap joints under different environmental conditions: deionized water, neutral saline solution and temperature/relative humidity controlled. In terms of fatigue strength, the water exposition promoted a significant effect but the saline solutions decreased significantly the fatigue life. The variation reached 39% (for105 cy-

cles) in relation to the control samples. The corrosion that occurred on the adherends was determinant on the fatigue failure mechanism as observed for the static tests. Finally, the stiffness monitored during the fatigue tests decreased with the number of cycles, evidencing the fatigue damage evolution. It was evident that the degradation process is faster in severe environments than for the control samples.

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