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Influence of low temperature cycles on the integrity of pretreated 2024 T3 aluminium joints by an ultrasonic method

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Many authors have considered the problem of adhesive bond durability over a long period of time or under special environmental conditions, and all have concluded that surface preparation is of paramount importance. Here, 2024 T3 aluminium alloy bonded joints, whose metallic plates underwent phosphoric acid pretreatment prior to bonding, are investigated before and after exposure to -40°C for 67 hours. This procedure of ageing is repeated several times and the samples are examined by ultrasonic Lamb waves after each cycle. In particular, the position of the cutoff-frequencies of ultrasonic guided waves which propagate into the bonded structure allows us to calculate the stiffnesses of the metal/adhesive interfaces via a rheological model. Thus, after exposure, a progressive decrease in the value of the stiffnesses is observed even for pretreated surfaces.

1 Introduction

Adhesively bonded joints are widely used in aerospace and automobile industries as a direct alternative to welding and riveting, because this technique avoids disadvantages such as for example, the loss of strength after welding or the danger of corrosion after riveting. The performance of an adhesive joint is measured by its capacity to sustain high loads under service conditions. In order to achieve durable joints, the metallic surfaces have to be pretreated prior to bonding.
For aluminium alloys, chemical surface pretreatments are often used, the most commonly being alkaline etching, acid pickling, and anodising. In particular, the phosphoric acid anodising creates a porous oxide coating at the surface of the metal, which provides protection against corrosion and exhibits a high resistance against humid or hot environment.

As far as the ageing of aluminium bonded joints is concerned, many authors deal with the durability of the joints after immersion in water or combination of high humidity and elevated temperatures.

In all the previous studies, the durability of the bonded joints is investigated by mechanical tests. On the other hand, many non-destructive techniques also allow to predict the state of a joint, and ultrasonics methods are the most promising. Althrough the ultrasonic characterization of the adhesive bonds is largely studied (Nagy and Adler [1], Heller *et al.* [2], Vlasie *et al.* [3]...), few authors are interested in the environmental degradation of the adhesion and many of the related works are mainly focused on ultrasonic imaging methods (Moidu *et al.* [4], Vine *et al.* [5], Rokhlin *et al.* [6]).

Here we develop a similar rheological model, which consists of replacing the two metal/adhesive interfaces by a uniform distribution of longitudinal and transversal springs. The stiffnesses of the springs depend on the pretreatment of the metallic surfaces prior to bonding, and here, we consider the phosphoric acid pickling. Our purpose is to study the durability of aluminium bonded joints after 67 hours periodic cycles at -40°C. This low temperature is used because in the aeronautic and aerospace industries, it is well-known that there are more and more bonded parts in the wings of the airplanes, in the nacelle structure and also in space vehicules. During the flight of commercial airplane, the exterior temperature is usually about $-50°$ C (at the altitude of $10km$). These considerations justify the temperature of cycling choosen in our study. Ultrasonic measurements are performed, at ambient temperature, after each ageing cycle and the behaviour of the resonances of the structure (Lamb modes) allows us to obtain some informations about the equivalent stiffnesses of the bonded interfaces. We observe that some Lamb modes are more sensitive than others to the effects of ageing.

2 Theory

Let us consider a tri-layer structure AA2024T3/ adhesive/ AA2024T3 (Fig. 1), for which the aluminium plates underwent a preliminary acid surface pretreatment. The contact between aluminium and adhesive is modelled by a surface distribution of longitudinal and transversal springs. We denote by K_L and K_T the surface stiffnesses of the springs, by ρ_1, c_{L1}, c_{T1} (respectively ρ_2, c_{L2}, c_{T2}) the mass density, the longitudinal and transversal wave velocities of the aluminium (respectively of the adhesive).

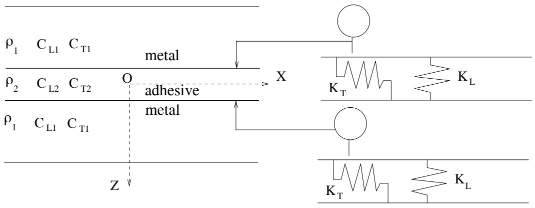

Figure 1: Geometry of the tri-layer structure.

In a recent paper (Vlasie *et al.* [8]), we show that the equations giving the cutoff-frequencies, noted f, of longitudinal modes are as follows:

$$\begin{aligned} &\mathrm{K}_L/(\rho_1 c_{L1})\Big[\cos(4\pi f h/c_{L1})\cos(\pi f d/c_{L2}) \\ &\text{-}\ \mathrm{z}_L\ \sin(4\pi f h/c_{L1})\sin(\pi f d/c_{L2})\Big] - \\ &\quad 2\ \pi f\sin(4\pi f h/c_{L1})\cos(\pi f d/c_{L2}) = 0, \end{aligned} \tag{1}$$

$$\begin{aligned} &\mathrm{K}_L/(\rho_1 c_{L1})\Big[\cos(4\pi f h/c_{L1})\sin(\pi f d/c_{L2}) \\ &+\ \mathrm{z}_L\ \sin(4\pi f h/c_{L1})\cos(\pi f d/c_{L2})\Big] - \\ &\quad 2\ \pi f\sin(4\pi f h/c_{L1})\sin(\pi f d/c_{L2}) = 0, \end{aligned} \tag{2}$$

where $z_L = (\rho_1 c_{L1})/(\rho_2 c_{L2})$, $2h$ is the thickness of the metallic plates, d is the thickness of the adhesive. The equations giving the transversal cutoff-frequencies are obtained by replacing the index L by T. These cutoff-frequencies are experimentally measured and allow the evaluation of the stiffnesses in order to obtain an estimation of the quality of the adhesion (see §3.3).

3 Experiment

3.1 Adhesive joints preparation and exposure

The metallic sheets used as adherends for the structural joints consist of 2024 $T3$ aluminium alloy ($110\ mm \times 140\ mm \times 3.95\ mm$). The mass density of aluminium is 2780 kg/m^3, and longitudinal and transversal wave velocities are 6330 m/s and 3140 m/s, respectively. The adhesive used for bonding is a bi-component epoxy (DGEBA/DDM) whose mass density is 1160 kg/m^3 and longitudinal and transversal velocities are 2527 m/s and 1356 m/s.
Before bonding with the adhesive, the aluminium sheets are pretreated with a solution of phosphoric acid and distilled water. Then, we made a mould with two aluminium plates, poured the liquid adhesive into this mould, and introduced the sample into an enclosure at 40° C for 24 hours so that the adhesive polymerized. The thickness of the epoxy resin is 450 μm. The total thickness of the samples is therefore 8.35 mm. Moreover, their length and width (140 mm and 110 mm) are large compared to the ultrasonic wavelengths so reflections on the edges of the structure can be neglected.

The adhesive joints are exposed at $-40°$ C for an arbitrary duration of 67 hours. The specimens are removed for the ultrasonic characterization, then reintroduced in the enclosure for another 67 hours cycle at $-40°$ C. This procedure is repeated four times and the cutoff-frequecies of the guided modes after each cycle are compared to those of the unexposed specimen. The cutoff-frequencies measured either on the cold samples, or after one day of exposure at ambient temperature have the same values. This means that the ageing introduces irreversible changes into the structure.

3.2 Experimental setup for the ultrasonic inspection

For the ultrasonic measurements of the adhesively bonded joints, we used the emission/reception technique. The ultrasonic signals are generated and received by a Panametrics transducer having a nominal frequency of 10 MHz. We are only focused on the study of the longitudinal cutoff-frequencies.

In Figs. 2 - 3, we plot the experimental resonance spectrum of the longitudinal waves for a single aluminium plate, and for an aluminium bonded structure respectively, when the metallic plates underwent an acid pickling pretreatment before joining. For values of the geometrical parameters $2h$ and d corresponding to industrial applications, we observe that the cutoff-frequencies of the bonded structure are duplicated around those of a single aluminium plate. This result was theoretically shown by Vlasie *et al.* [9].

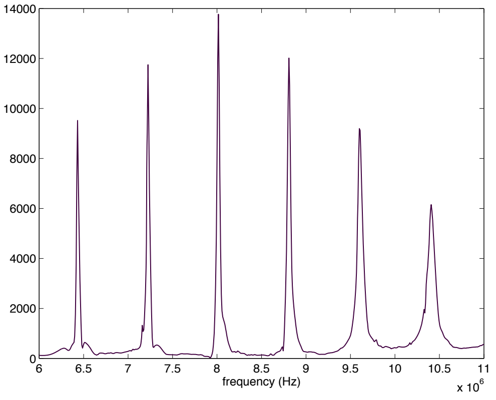

Figure 2: Resonance spectrum of the longitudinal waves for a plate.

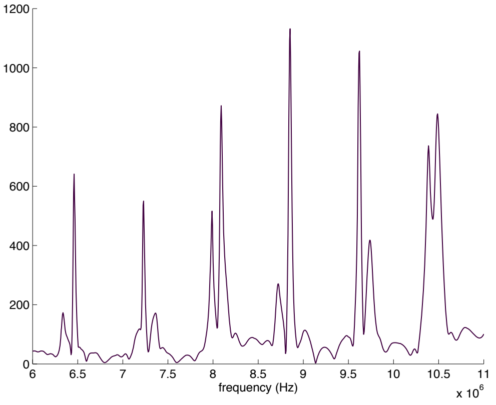

Figure 3: Resonance spectrum of the longitudinal waves for a bonded joint.

3.3 Results and discussion

All the resonance spectra of the longitudinal guided waves before and after the exposure to $-40°$C have the same shape as that of Figure 3. So, it is easy to compare the influence of the cycles of cooling on the values of the cutoff-frequencies. In this study, we investigate the frequency range between 6 MHz and 12 MHz. The measurements are carried out at twelve different points regularly distributed around the center of the sample (no reflection on the edges) and the results are homogeneous. Figure 4 compares the position of two resonances of longitudinal guided waves before (Cycle 0) and after exposure to $-40°\mathrm{C}$ (Cycles 1 to 4). These two resonances correspond to the duplication of a single aluminium plate resonance (indicated by the dotted vertical line).

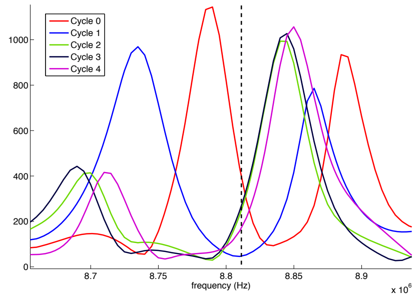

Figure 4: Evolution of the position of resonances during different cycles of ageing.

We observe that the cutoff-frequencies of the longitudinal modes reduce when the structure undergoes successive cycles of ageing. Moreover, if we study all modes between 6 and 12 MHz, we observe that some modes are more sensitive than others. These are the Lamb modes of the adhesive layer disturbed by the structure and, in this frequency range, they are located at 6.92 MHz and 10.24 MHz respectively, for the unexposed sample.

f^{C0}	f^{C1}	f^{C2}	f^{C3}	f^{C4}
8786	8729	8702	8690	8681
10240	9970	9802	9792	9750

Table 1: Evolution of some cutoff-frequencies (in kHz) according to the cycle of ageing

Table 1 presents the evolution of the experimental cutoff-frequencies of one aluminium mode and one adhesive mode going from the unexposed sample (f^{C0}) to the sample having undergone an exposure of 4 cycles of cold. For modes resulting from the duplication of an aluminium Lamb mode, the shift is smaller than 50 kHz, whereas the adhesive Lamb modes undergo a very significant shift after the first cycle (about 200 kHz). For the following cycles of ageing, the shift is smaller but it is always higher than the experimental error of the device, which is about 20 kHz.
These observations are not surprising because the adhesive is naturally more sensitive to the cooling than the aluminium. The Lamb modes of the adhesive are therefore more disturbed.

The question to be answered is how the adhesive layer could be disturbed?

First, the cold exposure of the sample and the fact that the two materials do not have the same thermal dilatation coefficient, produce a damage in the interfacial zone. To evaluate this possibility we calculate the corresponding stiffnesses using Eqs. (1) and (2) (see Table 2).

K_L^{V0}	K_L^{V1}	K_L^{V2}	K_L^{V3}	K_L^{V4}
4.84	3.03	2.67	2.53	2.42
45.1	20.8	14.9	14.5	13.1

Table 2: Calculated surface stiffnesses (in $10^{13} N/m^3$)

From the macroscopic point of view, the low temperature cycles induce a damage which can be observed in the decrease of stiffnesses values. So, according to the theory of damage we can conclude to a degradation of the bonded interfaces. However, we observe that the order of magnitude of the stiffnesses, in N/m^3, varies between 10^{13} and 10^{14}. However, these values correspond to good adhesion and, consequently, the five cycles of cold ageing did not produce a strong degradation.

Second, independently of adhesive degradation, the cooling introduces thermal prestress (more sensitive in the epoxy than in the aluminium) which produce micro-cracks in the epoxy layer. If the experimental wavelengths are small or of the order of the size of the micro-cracks, the modelling becomes complex because each micro-crack behaves like a screen in front of the acoustical waves; resulting a diffusion in all directions. On the other hand, for wavelengths larger than the size of the microcracks and from a macroscopic point of view, it is possible to introduce a homogeneous equivalent medium whose elasticity coefficients are smaller than those of the epoxy. In the frequency range considered in this paper ($6 - 12MHz$), the wavelengths in the epoxy layer vary between 200 and 400 μm. Here, the size of micro-cracks is 5 to 10 percent smaller than the wavelength and it is possible to define a homogeneous equivalent medium whose Young modulus decreases with the cycle of ageing and depends on the wavelength (the smaller is the wavelength, the more the ultrasonic waves perceive a large number of microcracks). Then longitudinal and transversal wave velocities and also the cutoff-frequencies decrease (even for constant values of stiffnesses). For instance, the longitudinal wave velocity in the state $C0$ is $2527m/s$, whereas in the state $C1$ the velocity is about $2389m/s$ near $7MHz$ and $2370m/s$ near $11MHz$.

The first explanation is attributed to adhesive degradation, the second one is attributed to a cohesive degradation of the epoxy, and both are complementary.

4 Conclusion

With regard to the perspectives of this work, two experimental studies should to be considered. First, it would be interesting to find the minimal time of ageing to which ultrasonic acoustic waves are sensitive. Some preliminary results show that ageing of the structure for less than 5 hours does not produce any effect on the cutoff-frequencies of the guided modes. Second, in the present study, the total duration of the ageing is 4 × 67 hours, i.e. 268 hours. It would be interesting to distribute this total duration differently in order to study the influence of the parameters "number of hours per cycle" and "number of cycles" on the frequencies of the guided Lamb waves.

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