ON THE EFFECT OF PRE-BOND MOISTURE IN THE ADHERENTS ON THE FRACTURE TOUGHNESS OF BONDED JOINTS FOR REPAIR PURPOSES

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Abstract

This study evaluates the sensitivity to pre-bond moisture of two adhesive films acting as bonding agents for composite repairs. To explore the effect of pre-bond moisture, the adherents were immersed in water at 70°C for 336 hr and then dried for 1 hr or 24 hr to lead to different moisture contents. Dry adherents were also used, as reference. Double Cantilever Beam specimens of bonded joints in "as-received" and "moisture saturated" conditions were tested under mode-I loading at room temperature and at 80ºC and 120ºC. The results show that pre-bond moisture has a dramatic effect on the fracture toughness values and on the failure mode. They emphasize the need of an exhaustive drying process prior to bond.

1 Introduction

Composite structures in service experience environmental and mechanical threats. The main environmental threats are related to the effect of temperature and moisture absorption as they can affect the strength of composite structures and reduce their lifetime. Therefore, moisture absorption should be taken into account during the structure design. In composite bonded joints, as those used for repairs, the amount of moisture uptake in the adherents, which might have an influence on the final performance of the joint, depends on a number of factors, e.g. specific adherent material used, exposure condition (temperature, humidity), exposure time and adherent thickness [1].

A few studies have been reported on the pre-bond moisture effect and most of them concluded that the presence of moisture in the composite lead to the reduction in joint strength [2, 3, 4, 5, 6]. There are some unexpected results for certain adhesive joints according to which the strength increased as the moisture level increased [2, 6, 7]. Matrix ductility caused by the plasticization of the matrix [5, 8, 9] are the possible reasons for the increase in G_{IC}. For certain adhesives, no significant changes occurred with increasing pre-bond moisture content up to certain level, above which it decreased [5, 7, 10]. No single theory or model exists with sufficient experimental support to explain the generalized relation between pre-bond moisture and the mechanical properties of joints. Because of the uncertainty on the effect of pre-bond moisture, repair procedures tend to prescribe a drying step before bonding the patch. A proper selection of the adhesive material and drying procedures to assure a low level of moisture would give positive influence on the performance of adhesive joints.

The objective of this study is to characterize the effect of pre-bond moisture on the Mode-I interlaminar fracture behavior of composite repairs. For pre-bond moisture study, the adherents were immersed in distilled water at 70°C for 336 hr and subsequently dried for 1 hr and 24 hr to attain different moisture levels. Bonded joints where then manufactured and distributed in three batches. One batch was tested "as-received" at room temperature. The other two batches of bonded joints were "moisture-saturated" and then tested at 80ºC and 120ºC. Mode I tests were performed by means of Double Cantilever Beam specimens.

2 Experimental Details

2.1 Materials

In order to determine the effect of pre-bond moisture on the fracture behavior of bonded composites joints, an experimental study was carried out with two different adhesive films denoted by F1 and F2. One of the adherents (a $[0, 90]_n$ plain weave carbon fabric epoxy prepreg) was previously cured in autoclave at 180°C and 700 kPa of applied pressure. The bonded joints with the pre-cured panel, the adhesive films (F1 and F2) and the fresh repair prepreg plies were cured in an oven under vacuum pressure. Time and temperature of the curing process was selected according to the manufacturer indications. A *teflon* insert was placed between the adherents so that an initial pre-crack of 60 mm was obtained. The dimensions of the panels were 350×300 mm and the specimens for Double Cantilever Beam fracture toughness tests were 25 mm in width and 145 mm in total length. Pre-cured panels were inspected by c-scan.

2.2 Pre-bond moisture uptake of the adherents

The immersion temperature (70ºC) and time (336 h) for the pre-cured adherents were selected because it was well-known that they cause a quick moisture uptake, leading to a situation close to equilibrium. The moisture substrate uptake, caused by this immersion process, was 1.6%. The choice of the drying conditions, in turn, obeyed the practical considerations of real repair operations. Drying for 24±0.5 hours at 80±5ºC is known to notably reduce the moisture content, although it does not remove it completely. It is a compromise between an acceptable reduction of moisture and an manageable duration of a repair intervention. The aim of the short drying cycle (1 hour at 80±5 ºC) was to investigate the tolerance of the different bonding agents to the cure in the presence of a considerable amount of moisture in the substrate. Assessing the feasibility of a reduced repair time was the final goal of this study. The moisture content before bonding was 1.25% and 0.33%, for the 1 hour and 24 hour drying at 80°C, respectively.

2.2 Conditioning of bonded joints

Once bonded joints were prepared, panels were cut to the desired specimen dimensions and distributed in 3 batches for each adhesive film, F1 and F2, (6 batches in total). Two of these batches were kept in a climatic chamber at 70°C and 85% RH condition to reach moisture saturation. Moisture content was monitored in accordance with the ASTM D-5229 standard

[12]. Specimens were removed from the climatic chamber after moisture equilibrium for testing. The specimens that were to be tested in "as received" conditions were stored in a desiccator box at room temperature.

2.3 Mechanical Testing

A universal testing machine, MTS Insight with a 1 kN load cell, at a crosshead rate of 5 mm/min was used for the DCB tests. Mechanical tests were performed according to ISO 15024 standard [11]. Test temperature for conditioned specimens until saturation (WET) was 80°C and 120°C, whereas as received specimens (AR) were tested at Room Temperature (RT). Tests were conducted in a laboratory controlled environment (23± 2°C, 50±5 RH) of the University of Girona **(**ISO 17025 and NADCAP "Non Metallic Material Testing" accredited). The crack length during propagation was monitored optically at the specimen's edge by means of a long distance microscope Questar QM100. Values of load, displacement, and crack length were measured simultaneously for crack initiation and crack propagation data according to ISO 15024 standard. The data was analyzed to determine the fracture toughness energy G_{IC} using several methods: Corrected Beam Theory, Modified Compliance Calibration and Area Method for crack onset and propagation

3 Results

3.1 Fracture toughness results (G_{IC})

Mean propagation values of G_{IC} calculated using the CBT method, are shown in Table 1. First of all, it should be highlighted, that a multiplicity of cracks was observed during testing of the joints without pre-bond moisture. In that case, the crack growth energy results from the contribution of every propagating crack, leading to larger values than those found if a single crack was propagating. Hence the mean propagation G_{IC} value measured for the joint without pre-bond moisture should be taken as a reference for comparison with the joints with pre-bond moisture only with caution.

The effect of the pre-bond moisture was clear at room temperature, RT, (Table 1): the pre-bond moisture in the adherents caused a decrease in the toughness of the resulting bonded joint (the drier the adherent, the higher the fracture toughness). Indeed, both adhesive film joints showed a significant (50%) increase in fracture toughness when the pre-bond moisture content was 0.33% in the substrate compared to 1.25%.

At higher temperature, for the "moisture-saturated" (WET) specimens, the effect of pre-bond moisture followed the same trend as for the as-received (AR) specimens tested at RT: the fracture toughness decreased with the pre-bond moisture content. Only F2 at 120ºC does not show a marked effect (no significant difference was observed between 1.25% and 0.33% pre-bond moisture content at this temperature). The effect of testing temperature (80ºC or 120ºC) on the G_{IC} values of the WET specimens was also clear: fracture toughness increased with the testing temperature irrespective of the pre-bond moisture content.

	Adherent pre-bond moisture content	Bonded joint condition	Fracture Toughness G_{IC} (J/m²)		
			Tested at R.T	Tested at 80°C	Tested at 120°C
Adhesive film F1	0.00%	AR	790	-	-
	1.25%	AR	409	-	-
	0.33%	AR	629	-	-
	0.00%	Wet (70°C/85 %RH)		718	**
	1.25%	Wet (70°C/85 %RH)		440	524
	0.33%	Wet (70°C/85 %RH)		554	1005
Adhesive film F2	0.00%	AR	804	-	-
	1.25%	AR	495	-	-
	0.33%	AR	751	-	-
	0.00%	Wet (70°C/85 %RH)	-	566	**
	1.25%	Wet (70°C/85 %RH)	-	336	443
	0.33%	Wet (70°C/85 %RH)	-	391	457

** Failed through the outer surface of the joints instead of interphase (see figure 5).

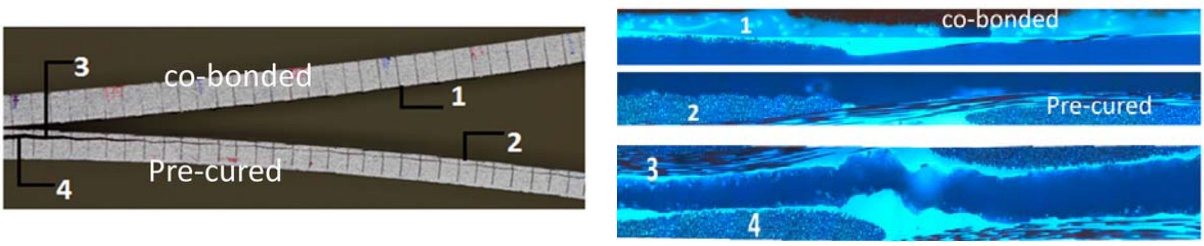
Table 1. Mean propagation values of fracture toughness (G_{IC}) resulting from the CBT data reduction method for the adhesive film bonded joints with and without pre-bond moisture

3.2 Fracture surface analysis

3.2.1 Testing at RT

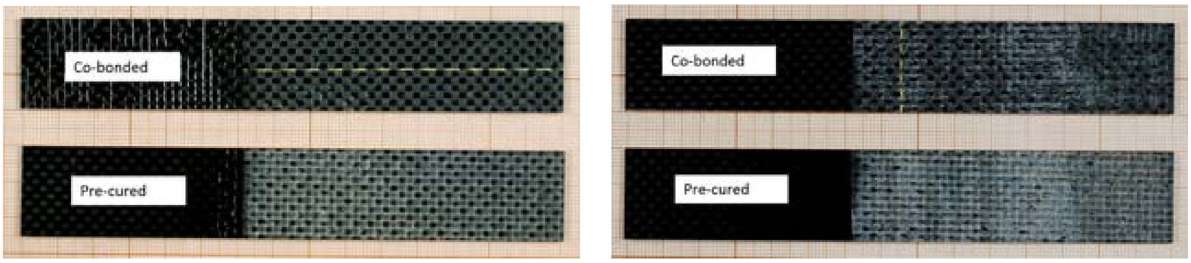
The fracture surfaces of "AR" and "WET" specimens were inspected visually and with optical microscope. In the figures, the adherents appearing on the top are the co-bonded adherents and those at the bottom are the pre-cured adherents. Multiple cracks were observed for the bonded joints with 0% pre-bond moisture for both adhesive films (F1 and F2) (Fig.1). It is clearly seen from the optical micrographies that the multiple cracks propagate in the pre-cured adherent itself. Multiple crack growth did not occur for the joint with pre-bond moisture (1.25% and 0.33%).

The failure mode changed dramatically for the two types of bonded joints (with both F1 and F2) when there was pre-bond moisture in the pre-cured adherent (Fig.2). Bonded joints with 1.25% pre-bond moisture, failed near the co-bonded adherent (Fig.3a). Same failure mode also occurred with little more contribution of cohesive failure for the joint with 0.33% pre-bond moisture (Fig.3b).



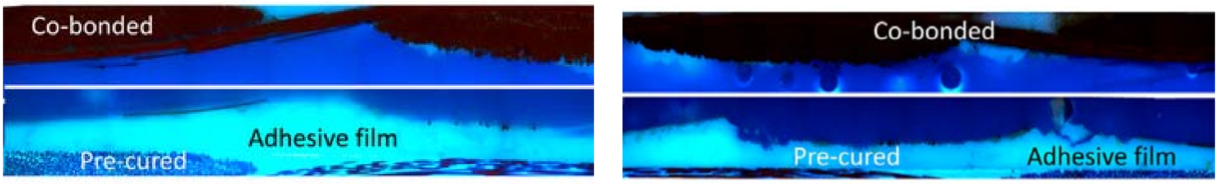
(a) Failure mode (b) Optical micrograph

Figure 1. Failure mode and optical micrograph of bonded joints using adhesive film F1 with 0% pre-bond moisture, tested at room temperature



(a) with pre-bond moisture (1.25%) (b) with pre-bond moisture (0.33%)

Figure 2. Fracture surface of bonded joints of adhesive film (F2) with different pre-bond moisture content (a) 1.25% (b) 0.33%, tested at room temperature



(a) with pre-bond moisture (1.25%) (b) with pre-bond moisture (0.33%)

Figure 3. Optical image of the transverse cross section of bonded joints using adhesive film (F2) with different pre-bond moisture (a) 1.25% (b) 0.33%, tested at room temperature

3.2.2 Results at 80°C and 120°C

Fig.4. shows the failure surface of a wet specimen tested at 80°C for the joints with adhesive film F2 without pre-bond moisture in the adherents. The surface shows a perfect cohesive failure of the adhesive. Optical microscopy confirmed this failure mode as it revealed adhesive film on both adherents. The same failure mode occurred for the bonded joints with adhesive film F1.

In the specimens of this type (no pre-bond moisture) tested at 120°C, the failure started from the outer surface of the co-bonded panel as shown in Fig.5. The test at 120°C had to be interrupted because of the failure of the adherent. This happened for the joints with the two adhesive films.

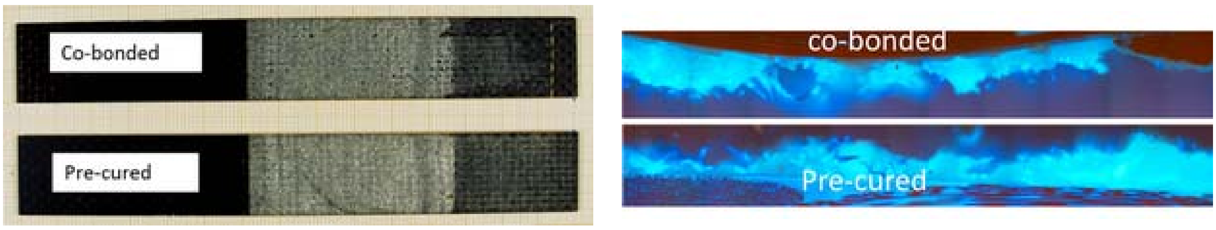


Figure 4. Failure surface and optical micrograph of bonded joints using adhesive film (F2) with 0% pre-bond moisture, tested at 80°C

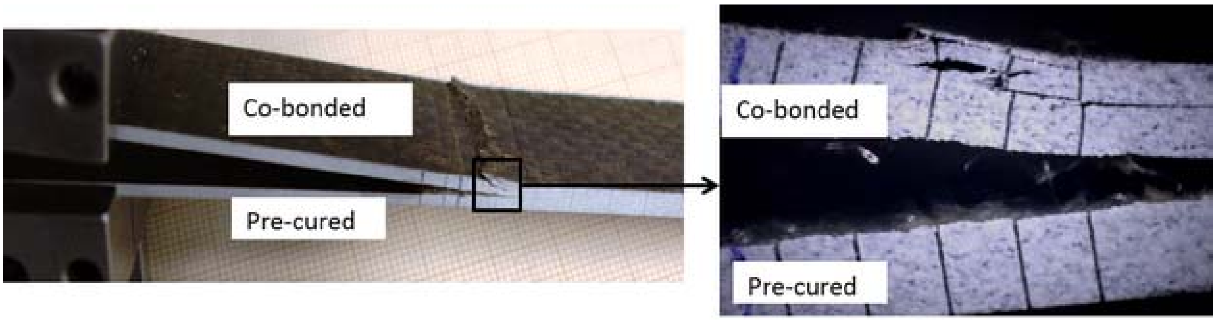
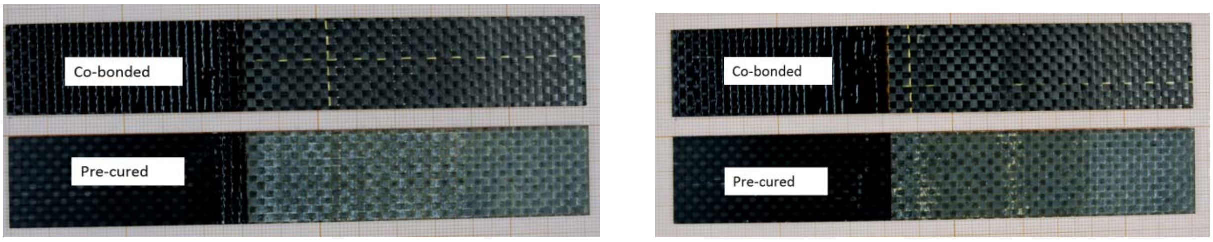


Figure 5. Failure mode of bonded joints using adhesive film (F1) with 0% pre-bond moisture, tested at 120°C

The pre-cured adherent was completely covered with adhesive film, as shown in Fig.6, for both adhesive films in all pre-bond moisture conditions tested at 80°C and 120°C. From the optical microscopy images it is revealed that failure mostly took place in between the adhesive film and the co-bonded adherent (Fig.9). The same failure mode occurred at 80°C and 120°C for both pre-bond moisture level (0.33% and 1.25%).



(a) 80°C, 1.25% pre-bond moisture (b) 120°C, 1.25% pre-bond moisture

Figure 6. Fracture surface of joints with 1.25% pre-bond moisture bonded using adhesive films tested at (a) 80°C and (b) 120°C

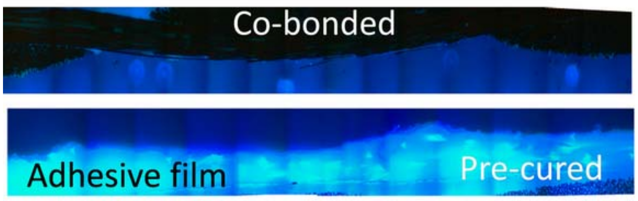


Figure 7. Optical image of the transverse cross section of bonded joints using adhesive film (F1) with 1.25 % pre-bond moisture, tested at 120ºC

4 Conclusions

Co-bonded joints with two adhesive films acting as bonding agents cured under vacuum pressure for in-service repairs were produced. The effect of moisture uptake in the adherents before bonding and the effect of test temperature on wet specimens were analyzed. The main outcomes from of the present study are as follows:

Joints with 0% pre-bond moisture tested at room temperature exhibited multiple cracks in the pre-cured adherent in addition to the insert plane, indicating that the bond joint is stronger than the adherent itself. However, the presence of pre-bond moisture in the pre-cured adherent produced a remarkable change in the failure mode, with the crack propagating in the interface between the adhesive film and the co-bonded adherent. This wet-wet failure was more evident in the samples with high pre-bond moisture content (1.25%). Samples with lower pre-bond moisture (0.33%) exhibited a slight contribution of cohesive failure.

Extending the duration of the drying operation of the substrate with pre-bond moisture for the joints with adhesive films, F1 and F2, showed a positive increment in fracture toughness. To that purpose, more exhaustive drying processes might be necessary: while enlarging significantly the drying time might not be practical, increasing the drying temperature is still suitable.

Even though both studied adhesives, F1 and F2, presented the same failure pattern, the fracture energy at room temperature of the adhesive film F2 was almost 20% higher than that of the adhesive F1 for all pre-bond moisture conditions.

0% pre-bond moisture conditioned joints showed also higher fracture toughness compared to the 1.25% and 0.33% pre-bond moisture joints at 80°C. Matrix deformation and cohesive failure occurred and these are the possible explanations for the increment in fracture toughness. When comparing the two adhesive films tested in these conditions, F1 presented higher fracture toughness than F2 at all pre-bond moisture contents, reversing the trend observed at room temperature. F1, having lower values at room temperature than F2, improved its toughness after being wetted and tested at 80ºC, which seems to indicate a higher plastic deformation in this adhesive film.

At high test temperature, 120°C, with no pre-bond moisture, the co-bonded adherent in the wet specimens appeared to be the weakest part of the system, suffering multiple cracks away from the bond joint.

In the presence of pre-bond moisture, the interface between the co-bonded adherent and the adhesive films weakened in the wet specimens tested at both 80ºC and 120ºC. The adhesive films remained bonded in all cases to the pre-cured adherent. This wet-wet failure had been also observed in the specimens with pre-bond moisture tested at room temperature.

These results indicate the need of a careful and detailed characterization of the bonded joints, paying attention not only to the fracture toughness values but also to the failure made. For the repair systems studied here the presence of pre-bond moisture has a dramatic effect on the failure mode, moving it from the adherents themselves or the bulk adhesive films to the wet-wet interface in all cases, whatever the subsequent aging of the bonded joint and the test temperature. To avoid this failure mode, the removal of moisture from the pre-cured adherent appears to be essential.

Once the impact of the pre-bond moisture has been eliminated by the proper drying process, careful choice of the adhesive and the repair prepreg and consideration of the operating conditions and in-service temperature should not be ignored, as evidenced by the comparison of the different behaviour of the bonded joints with F1 and F2 at room temperature/as received versus 80ºC/wet and 120ºC/wet.

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