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TECHNICAL PAPER

Effect of moisture on the adhesion of CFRP-to-steel bonded joints using peel tests

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Abstract
In this work, an experimental study has been carried out to evaluate the effect of salt water condition on the long-term adhesion of composite-to-metal bonded joints using peel tests. A new test configuration is tested via fully and partially immersed specimens with 500 mm length. Fracture surfaces of non-aged samples exhibited a cohesive failure within the adhesive layer, which indicates a good adhesion of the joint. Results revealed a significant difference in the interface adhesion between aged and non-aged condition after 150 days of immersion. Partially immersed specimens allow to evaluate the adhesion performance of the joint under dry and wet condition in single specimen using a simple peel test.

Keywords Adhesively bonded joints · Peel tests · Moisture · CFRP · Salt water

1 Introduction

Nowadays, composite materials are widely used for repair of metallic components in offshore, onshore, oil and gas industries. Adhesive bonding is the most efficient joining technology in terms of weight and performance to join these two materials. Therefore, composite-to-metal bonded joints are nowadays a common practice in such maritime industries. The effect of multimaterial bonding process on joint strength has been investigated by many researchers and found a positive influence on the performance of bonded structure [1–3]. However, limited research is found on the performance of such hybrid joints under the harsh maritime environments. One of the most critical aspects for the long-term durability of bonded joints is the adhesion at the interfaces. With the increasing use of composite-to-metal bonded joints, attention should be paid on the adhesion quality in the presence of environmental effect to assure long-term durability of the multimaterial bonded joints.

During service, metal structure can be exposed to extreme environmental condition, which impacts their performance. This can be especially severe in offshore and onshore unit, where the metal pipe is always in contact with the salt water [4, 5]. Several works were carried out to assess the influence of environmental parameters on the durability of bonded structures [5–9]. There are many studies carried out on the ageing effect, using the composite–composite bonded joints (mode I, mode II and mixed mode) and only very few carried out with the composite–metal bonded joints [10–14]. Most of the results found a detrimental effect of moisture on the performance of the bonded joints and show that moisture is one of the most problematic factors, when it comes to the durability of adhesive joints with FRP and metallic adherents [15–18]. Salt water immersion is one of the worst ageing conditions to affect the joint performance, because not only the adhesion is affected by the moisture but also the metal gets corrode easily. So, it is of prime importance to assess the interface adhesion between the metal–composite joint in the presence of different environmental conditions.

Interface adhesion is one of the most important parameters for assuring the integrity of composite–metal bonded joints. Peel test is a suitable destructive test method to examine the interface adhesion properties, while other test

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methods such as single leg bending (SLB) and double lap joint (DLJ) examine the mechanical properties of the joint rather than the interface adhesion. The floating roller peel test is one of the most commonly used methods in metal bonding because it is a simple, fast and reliable test method to assess adhesion [19–23]. For metal substrates, the peel test is well developed and documented. Nevertheless, few research has been done on this type of test when applied to dissimilar substrate joints [24, 25]. Some researchers [26–28] performed peel tests on composite–metal bonded joints and they successfully assessed the interface adhesion between composite–metal joints. A simple and straightforward test coupon is needed to assess the adhesion of metal–composite bonding and evaluates the adhesion interface of interest.

In this study, floating roller peel tests are used to assess the adhesion properties of the composite–metal bonded joints. A new method of analysis is proposed in which long peel specimens are used to evaluate the effect of ageing and non-ageing in the same specimen. This work aims to evaluate the effect of salt water on the adhesion of carbon fibre reinforced polymer (CFRP)-to-carbon steel bonded joints using peel tests and to evaluate the new method of analysis proposed.

2 Experimental methods

2.1 Materials

Normally, steel pipes are used to carry fluid over a long distance because of its good mechanical properties [29, 30]. To assess the interface adhesion between the existing steel structure and the CFRP repair, peel test specimens were manufactured with carbon steel and CFRP substrates. The peel specimens consisted of a CFRP rigid adherend bonded to a carbon steel flexible adherend. An NVT201E® epoxy resin (Novatec Ltd., Rio de Janeiro, Brazil) was used as an adhesive to join the substrates. Carbon fibre fabrics with a density of 430 g/m^2 were selected to make the composite adherend. Each fabric is composed of two unidirectional laminas of continuous fibres oriented perpendicula [0/90]. PIPEFIX® epoxy resin (Novatec Ltd., Rio de Janeiro, Brazil) was selected for impregnation of the composite laminate. A 0.4 mm thick carbon steel ASTM A36 plate was used as a flexible adherend.

2.2 Sample preparation

The composite-to-metal bonded joint test specimens were manufactured by the same process used in composite patch repairs of oil and gas production platforms [29, 30]. The carbon steel plate was blasted with G-40 steel shot and then degreased using acetone. The final surface roughness measured was 12.26 µm root mean square deviation (Rq) and 76.12 µm total height of the roughness profile (Rt). The epoxy resin NVT201E® was then applied over the treated steel plate surface. Afterwards, the lamination process was started by alternating application of carbon fibre fabric layers and the impregnation resin PIPEFIX®, as shown in Fig. 1. Six plies of carbon fibre were used to produce the composite with a final layup of $[0/90]_6$. This means that the CFRP is oriented alternatively at the specimen longitudinal and transverse direction. The curing process was performed at room temperature for 2 h. One layer of glass fibre dry chopped strand mat (300 g/m^2) was used between epoxy adhesive and carbon fibre fabric layer to avoid direct contact between carbon fibre and steel plate. The mechanical properties of the composite laminate and adhesive were determined in a previous work [28].

Peel test specimens were manufactured with 550 mm length flexible substrate, 500 mm length rigid substrate and 25 mm width. The rigid substrate is made of carbon fibre laminated composite, and the flexible substrate is made of carbon steel, as shown in Fig. 2.

The salt water ageing process was employed to evaluate the long-term effect of environmental exposure on the joint. The specimens were kept inside the salt water tank (5% NaCl) at room temperature for 40 days and 150 days. The ageing process simulates the effect of environmental conditions, wherein the bonded repairs are exposed to marine environment. Specimens were exposed to ageing in two different conditions: partially immersed in salt water, i.e. 50% of bonding length was in salt water and remaining in atmospheric conditions; and fully immersed in salt water. Some reference specimens were tested without any ageing. Figure 3 shows the specimens in the salt water tank and Table 1 provides the ageing conditions tested and the number of specimens for each condition.

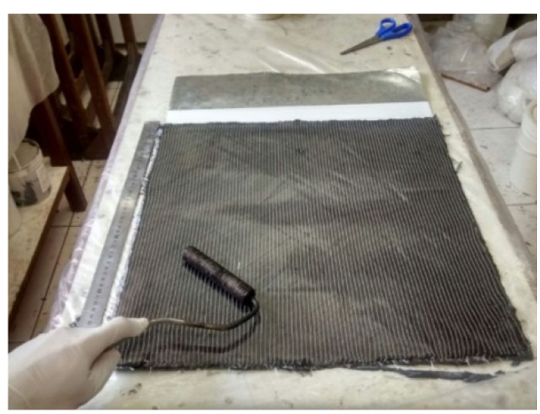


Fig. 1 Carbon fibre lamination process

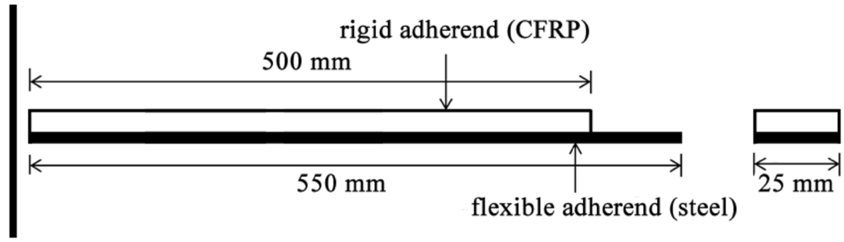


Fig. 2 Peel test specimen

Fig. 3 Specimens in salt water tank

Table 1 Specimens tested

Condition	Number of specimens
Non-aged	3
40 days fully aged	3
40 days partially aged	3
150 days fully aged	3
150 partially aged	3

2.3 Floating roller peel tests

The floating roller peel test allows determining the peel strength between a rigid and a flexible adherend. Tests were performed on the non-aged and aged CFRP-to-steel specimens at room conditions, by means of an electro mechanic Instron machine (Series 5966) with a maximum capacity of 20 kN, coupled with a load cell of 1 kN. The testing speed was 152 mm/min, according to ASTM D-3167 for metal substrates [31]. During the test, the flexible adherend is peeled off from the rigid adherend. Figure 4 shows the test setup. Load–displacement curves were recorded during testing.

3 Results and discussion

3.1 Moisture absorption

The specimens were immersed in salt water for different duration to attain different moisture levels in the joints. The specimens were periodically removed from the tank, wipe with tissue paper and then weighed. Weighing was performed on a weight scale with an accuracy of 0.1 mg. Representative curves of the moisture absorption of fully and partially immersed specimen are shown in Fig. 5.

Mass uptakes of 3.7 and 4.2% were recorded in the fully immersed specimen after 40 days and 150 days of ageing, respectively. In case of partially immersed specimens, moisture uptake of 2.4 and 2.8% was recorded for the same periods. As it can be seen from Fig. 5, both curves show similar behaviour of fully and partially immersed specimens. The moisture absorption is higher compared to similar studies of carbon–epoxy laminates [12, 17] and the most possible reason for this is the manufacture process of the composite used in this work (hand layup may produce voids inside the composite that promotes moisture penetration). Furthermore, most of the moisture absorption occurred in the measurement range between 20 and 27 days in both cases. The presence of moisture causes plasticization, swelling, and degradation of the polymer matrix and deteriorates the fibre/matrix interface [32, 33]. These effects may have weakened the composite properties during the first weeks. This combined with the presence of voids in the material might have caused the steep mass uptake observed.

Figure 6 shows the ageing effect on the composite and steel adherend materials. The composite adherend is shown before immersion (Fig. 6a) and after 150 days of exposure to salt water (Fig. 6b). Carbon steel is also shown before immersion (Fig. 6c) and after the same period of exposure (Fig. 6d). The effect of corrosion along the steel surface can be visualized when comparing the carbon steel surface before and after conditioning. Otherwise, the composite surface was merely covered with salt in the same period. This exemplifies one of the main advantages of composite materials in relation to metals on harsh environment.

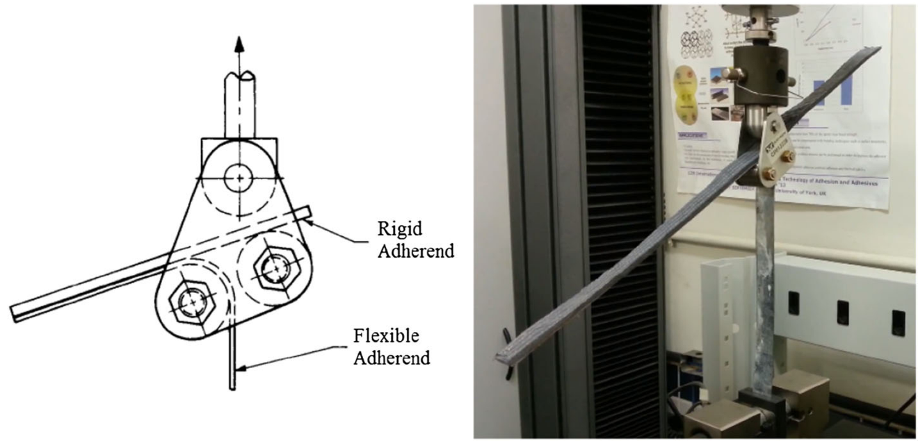


Fig. 4 Floating roller peel test set up

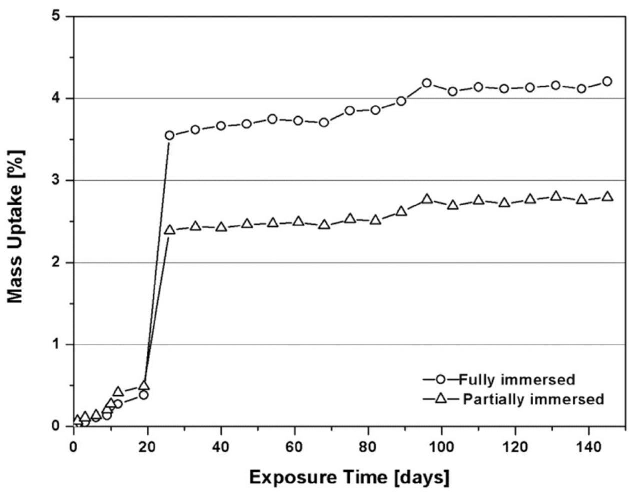


Fig. 5 Moisture absorption by the partially and fully immersed specimens

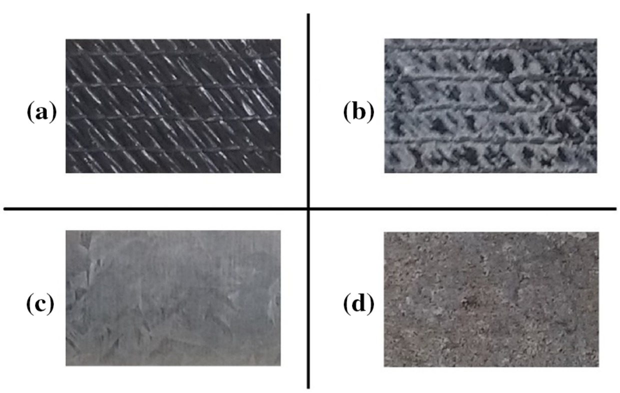


Fig. 6 Ageing effect on composite (**a** and **b**, before and after immersion, respectively) and metal adherends (**c** and **d**, before and after immersion, respectively)

3.2 Peel loads

Representative curves of the test results on fully immersed specimens and non-aged representative specimens (dry) are shown in Fig. 7. The average peel loads (F_{ave}) of fully immersed specimens were determined over the range of 25–400 mm of displacement. The values are shown in Table 2 as average and standard deviation of the three tests performed in each condition. The large dispersion in the results, shown by the high value of standard deviation, makes it difficult to analyse the ageing in 500 mm length joints.

Representative load–displacement curves of the test on partially immersed specimens are shown in Fig. 8. The region between 0 and 250 mm displacement corresponds to the aged portion of the specimen and the remaining length corresponds to the non-aged portion. The reference non-aged specimen curve (dry) is also shown.

The average peel loads of partially immersed specimens were determined over a 150 mm length in two different intervals: 25–175 mm and 250–400 mm. These intervals were chosen to separately analyse the aged and non-aged parts of the specimen. The results are shown in Table 3, as average and standard deviation of the three tests performed

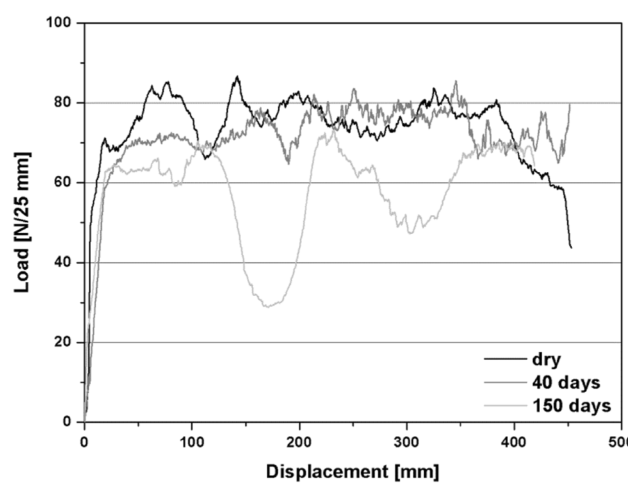


Fig. 7 Representative load –displacement curves of dry and fully aged specimens (40 days and 150 days)

Table 2 Average peel loads of fully immersed specimens

Specimen	F_{ave} (N/25 mm)
Non-aged	63.2 ± 11.8
40 days aged	69.9 ± 3.5
150 days aged	63.6 ± 14.1

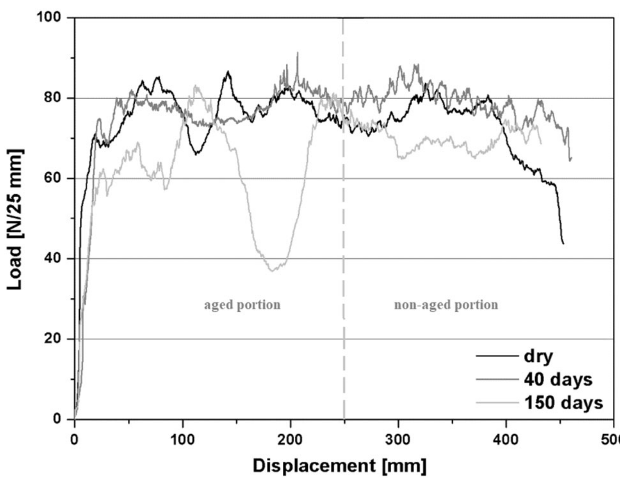


Fig. 8 Representative curves of dry and partially aged specimens (40 days and 150 days)

Table 3 Average peel loads of partially immersed specimens

Specimen	F_{ave} (N/25 mm)	
40 days aged	Non-aged	78.0 ± 6.9
	Aged	71.6 ± 8.1
150 days aged	Non-aged	64.0 ± 9.2
	Aged	62.0 ± 6.1

in each condition. The average peel loads are lower in the aged part for both 40 and 150 days of ageing, as expected. Significant difference in peel load is observed between 40 days and 150 days for ageing condition. The ageing effect in peel loads is noted on a single partially aged test specimen.

Load–displacement curves of 40 days aged specimens exhibit the same pattern in both fully and partially aged conditions. It confirms that the influence of moisture on the interface adhesion is insignificant after 40 days immersed in salt water. Otherwise, large decreases in peel loads are observed in fully immersed specimens in salt water for 150 days (Fig. 7). The same pattern is noted in the aged portion of partially immersed specimens for the same period (Fig. 8). This evidences the effect of ageing on the test results. High deviations on the average peel loads imply in longer ageing periods for more evident results.

3.3 Fracture Surfaces

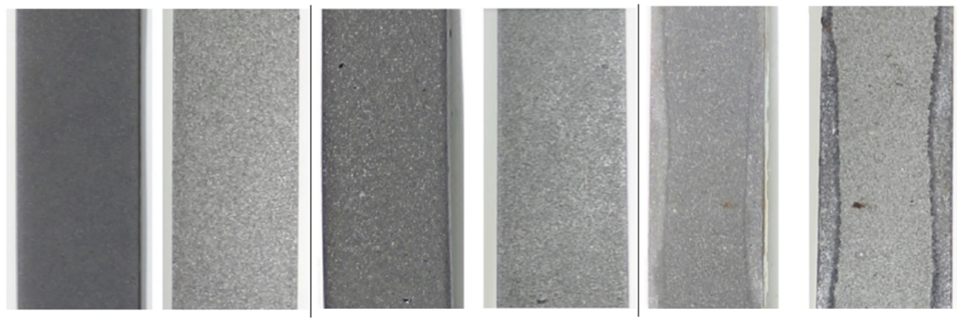
Visual inspection of the substrates fracture surfaces provides valuable information about the nature of the failure mode. Figure 9 shows the fracture surfaces of the composite adherend (left) and steel adherend (right) of non-aged and aged specimens. Cohesive failure is identified as the prevailing failure mode for non-ageing specimens (Fig. 9a). This indicates a good adhesion quality between the composite and steel substrates. It is also observed that the failure occurs near the interface between the steel (flexible adherend) and the adhesive, suggesting the presence of interfacial failure. The fracture surfaces of a 40-day aged specimen are shown in Fig. 9b. In this case, cohesive failure is also identified as the main failure mode, with a small influence of the moisture in the adhesive layer (less than 5% of the surface). This shows that the adhesive layer is not significantly affected after 40 days immersed in salt water. Figure 9c shows the fracture surfaces of a 150-day aged specimen. It is noted that after 150 days of exposure the adhesive layer was significantly affected by moisture (about 20% of the surface). The moisture effect pattern remains at the edges of the surfaces.

Moisture absorption (Fig. 5) and fracture surfaces (Fig. 9) are now related. Moisture was mostly absorbed by the composite substrate during the first 40 days of ageing. The adhesive layer was not significantly affected until this moment. It is also observed that there was a considerable growth of the region of the fracture surface affected by moisture between 40 and 150 days of ageing. This shows that most of the moisture absorption in the adhesive layer occurred during this period. The pattern of fracture surfaces at different ageing times shows that moisture penetrates from the edges toward the centre of the adhesive layer. It implies that a reduction of the specimen width can enhance the results related to the conditioning process. A reduced width would increase the ratio of affected area on the fracture surface. Finally, the white colouration presented in areas affected by moisture suggests that there was deposition of salt at the adhesive layer during ageing times.

4 Discussion

The average peel load of 63.2 ± 11.8 N/25 mm was measured for non-aged specimens (500 mm length) tested in the present work. While in previous work [28], similar specimens with 250 mm length were tested and an average peel load of 73.6 ± 1.2 N/25 mm was determined along 150 mm length. Increasing the length of the specimen

Fig. 9 Fracture surfaces of **a** non-aged, **b** 40 days aged and **c** 150 days aged specimens after peel tests



promoted an intensification in the discrepancies of test results. This may be caused by an increase in defects originated from specimens manufacture. Moreover, the salt spray ageing process used in [28] generated consistent results in 90 days while the same results were not reached in 150 days ageing in salt water.

To obtain a better understanding of the ageing effects on the adhesion performance, peel loads were related to fracture surfaces of 150 days aged joints, as they presented a more significant influence of moisture on the adhesive layer. Figure 10 shows a comparison between load–displacement curve and fractured surface of the flexible substrate in a partially aged tested specimen.

Three different regions can be identified by visual inspection along the curve: I, II and III. Regions I and II correspond to the aged part of the specimen while region III is the non-aged part. The moisture penetration at the fracture surface is small and reasonably uniform in the region I (between 25 and 145 mm of displacement) and corresponds to an average peel load of 62.6 ± 12.1 N. This value is close to the average peel load of 150 days fully immersed specimens showed in Table 2. Region II presents a considerable increase of moisture penetration, which is followed by a decrease in peel loads until 36.8 N. Region III (from 220 mm to the end) presents an average peel load of 71.1 ± 3.8 N. This value is close to the average peel load of non-aged specimens (Table 1). There is a 12% reduction in the average peel load between regions I and III. In addition, the minimum peel load occurred in region II is about 48% lower than the average peel load of the non-aged region. These percentages are proportional to the penetration of moisture in the fracture surface. This reveals the relationship between ageing effect and reduction of adhesion quality.

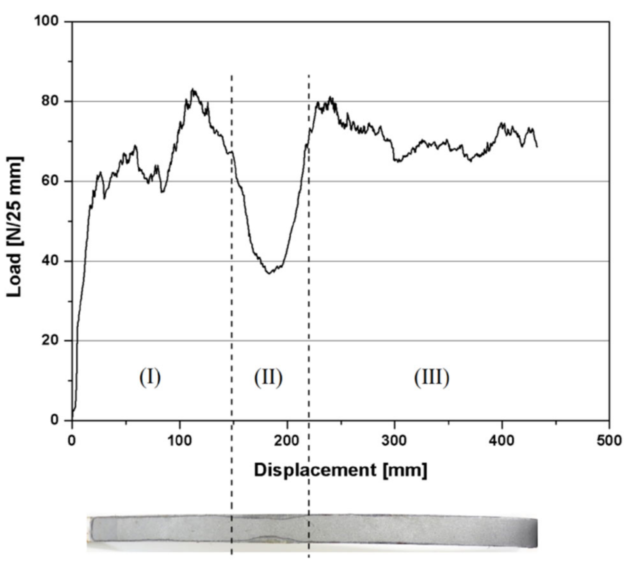


Fig. 10 Comparison between peel load and fracture surface of 150 days partially aged specimen

5 Conclusions

Peel tests were carried out on composite-to-steel bonded joints to study the effect of salt water. A recent developed type of peel test was applied in which a composite rigid adherend is bonded to a flexible metal adherend. New specimen configurations with longer length (500 mm) were fabricated to study the wet (ageing) and dry (non-ageing) behaviour in the same specimen.

The following conclusions can be drawn:

- There is no significant effect of moisture on the adhesion after 40 days of immersion in salt water. Fracture surface shows a cohesive failure mode as in the non-aged specimens. During this period, the moisture absorption by the adhesive interface is minimal.
- There is a considerable gain in moisture at the interface between the immersion period of 40 days to 150 days. However, the ageing process proved to be time-consuming. For faster and more evident results, either the specimen width should be reduced or a more severe ageing process should be employed (at higher temperatures for example).

- The penetration of moisture at the adhesive interface always occurs from the edges towards the centre of the interface, presenting an irregular pattern. Longer ageing periods produce higher moisture penetrations and therefore lower peel strengths.
- The peel strength of the non-aged specimens changed according to the total length of the specimens. Increasing the length of the specimen intensifies the discrepancies of the test results. This may be caused by an increase in intrinsic manufacture defects of the specimens. However, the new configuration shows a more realistic situation where the composite repairs are applied on larger area.
- The new specimen configuration (500 mm length) successfully assessed the interface adhesion for ageing and non-ageing condition in same specimen using peel test, provided the defect-free manufacturing of the composite–metal bonded joints.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

References

1. Teixeira de Freitas S, Sinke J (2015) Failure analysis of adhesively-bonded skin-to-stiffener joints: metal–metal vs. composite-metal. Eng Fail Anal 56:2–13
2. Arenas JM, Alía C, Narbón JJ, Ocanã R, González C (2013) Considerations for the industrial application of structural adhesive joints in the aluminium-composite material bonding. Compos Part B-Eng 44:417–423
3. Rudawska A (2010) Adhesive joint strength of hybrid assemblies: titanium sheet-composites and aluminium sheet-composites—experimental and numerical verification. Int J Adhes Adhes 30(7):574–582
4. Heshmati M, Haghani R, Al-Emrani M (2016) Effects of moisture on the long-term performance of adhesively bonded FRP/steel joints used in bridges. Compos Part B-Eng 92:447–462
5. Karbhari VM, Chin JW, Hunston D, Benmokrane B, Juska T, Morgan R, Lesko JJ, Sorathia U, Reynaud D (2003) Durability gap analysis for fiber-reinforced polymer composites in civil infrastructure. J Compos Constr 7:238–247
6. Boer P, Holliday L, Kang THK (2014) Interaction of environmental factors on fiber reinforced polymer composites and their inspection and maintenance: a review. Constr Build Mater 50:209–218
7. Karbhari VM (2014) Rehabilitation of metallic civil infrastructure using fiber reinforced polymer (FRP) composites: a materials and systems overview at the adhesive bond level. Woodhead Publishing, Sawston
8. Boer P, Holliday L, Kang THK (2013) Independent environmental effects on durability of fiber reinforced polymer wraps in civil applications: a review. Constr Build Mater 48:360–370
9. Karbhari VM (2007) Durability of composites for civil structural applications. Woodhead Publishing, Sawston
10. Fernandes RL, de Moura M, Moreira RDF (2016) Effect of moisture on pure mode I and II fracture behaviour of composite bonded joints. Int J Adhes Adhes 68:30–38
11. Han X, Crocombe AD, Anwar SNR, Hu P (2014) The strength prediction of adhesive single lap joints exposed to long term loading in a hostile environment. Int J Adhes Adhes 55:1–11
12. Liu SF, Cheng XQ, Zhang Q, Zhang J, Bao JW, Guo X (2016) An investigation of hygrothermal effects on adhesive materials and double lap shear joints of CFRP composite laminates. Compos Part B-Eng 91:431–440
13. Papanicolaou GC, Charitidis P, Mouzakis DE, Karachalios E, Jiga G, Portan DV (2016) Experimental and numerical investigation of balanced Boron/Epoxy single lap joints subjected to salt spray aging. Int J Adhes Adhes 68:9–18
14. Halliday ST, Banks WM, Pethrick RA (1999) Influence of humidity on the durability of adhesively bonded aluminium composite structures. Proc Inst Mech Eng Part L 213:27–35
15. Owens J, Lee-Sullivan P (2000) Stiffness behaviour due to fracture in adhesively bonded composite-to-aluminum joints II. Experimental. Int J Adhes Adhes 20(1):47–58
16. Han X, Crocombe AD, Anwar SNR, Hu P (2014) The strength prediction of adhesive single lap joints exposed to long term loading in a hostile environment. Int J Adhes Adhes 55:1–11
17. Nguyen T, Bai Y, Zhao X, Al-Mahaidi R (2012) Durability of steel/CFRP double strap joints exposed to sea water, cyclic temperature and humidity. Compos Struct 94:1834–1845
18. Mukhopadhyayau P, Swamy RN, Lynsdale CJ (1998) Influence of aggressive exposure conditions on the behavior of adhesive bonded concrete GFRP joints. Constr Build Mater 12:427–446
19. Sargent JP (2005) Durability studies for aerospace applications using peel and wedge tests. Int J Adhes Adhes 25:247–256
20. Kawashita LF, Moore DR, Williams JG (2004) The development of a mandrel peel test for the measurement of adhesive fracture toughness of epoxy–metal laminates. J Adhes 80:147–167
21. Wei Y, Hutchinson J (1998) Interface strength, work of adhesion and plasticity in the peel test. Int J Fract 93(1–4):315–333
22. Kim J, Kim KS, Kim YH (1989) Mechanical effects in peel adhesion test. J Adhes Sci Technol 3(1):175–187
23. Crocombe AD, Adams RD (1982) An elasto-plastic investigation of the peel test. J Adhes 13:241–267
24. Teixeira de Freitas S, Sinke J (2015) Test method to assess the interface adhesion in composite bonding. J Appl Adhes Sci 3:1–9
25. Hart-Smith LJ (1999) A peel-type durability test coupon to assess interfaces in bonded, co-bonded, and co-cured composite structures. Int J Adhes Adhes 19:181–191
26. Kawashita LF, Kinloch AJ, Moore DR, Williams JG (2006) A critical investigation of the use of a mandrel peel method for the determination of adhesive fracture toughness of metal-polymer laminates. Eng Fract Mech 73:2304–2323
27. Teixeira de Freitas S, Sinke J (2014) Adhesion Properties of Bonded Composite-to-Aluminium Joints Using Peel Tests. J Adhes 90:511–525
28. Teixeira de Freitas S, Banea MD, Budhe S, de Barros S (2017) Interface adhesion assessment of composite-to-metal bonded joints under salt spray conditions using peel tests. Comp Struct 164:68–75
29. Meniconi LCM, Porciúncula IN, McGeorge D, Pedersen A (2010) Structural repair at a production platform by means of a composite material patch. In the proceeding of the Offshore Technology Conference. https://doi.org/10.4043/20657-ms

30. Meniconi LCM, Lana LDM, Morikana SRK (2014) Experimental fatigue evaluation of the composite repair of a metallic ship hull. App Adhes Sci 2(27):1–13
31. ASTM D3167-10 (2010) Standard test method for floating roller peel resistance of adhesives
32. Banea MD, Silva LFM (2009) Adhesively bonded joints in composite materials—an overview. Proc IME J Mate Des Appl 223:1–18
33. Budhe S, Banea MD, de Barros S, da Silva LFM (2017) An updated review of adhesively bonded joints in composite materials. Int J Adhes Adhes 72:30–42