Effect of adhesive thickness on the strength of steel-composite joints aged in wet environment

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Abstract. This paper presents the effect of adhesive layer thickness on the static strength of adhesively bonded steel-glass fiber-reinforced unsaturated polyester resin (GFRP) lap joints after being aged in water at a temperature of 60°C for 15 days. The adhesive thickness was varied from 0.1 mm to 0.5 mm. It was found that for the dry joints, the static strength increased with the increase of adhesive thickness. However, in wet condition, water really had a significant detrimental effect on the strength of the joints, as most of the strengths of wet joints were much lower than those of the dry joints. In dry condition, mode of failure changed from interfacial (at the adhesive thickness of 0.1 mm) to mix failure; however, for the wet joints; most of failures were at the interface of adhesive/steel.

1. Introduction

Nowadays, adhesively bonded metal-composites joints gain more applications in transportation industry in order to develop lightweight vehicles. Other sectors used adhesively bonded joints technology, mainly for strengthening steel-made bridge structures, concrete and wood structures. The drawback in making use of adhesively bonded joints is the capability of the adhesive to absorb water from the surrounding environment. It is well known that the absorbed water has deleterious effect on the strength of the adhesive joints; therefore, designing an adhesive joint in those applications become attractive researches, considering the effect of adhesive thickness, type of adhesive and substrate, and type of environment.

Theoretically, (i.e. Goland-Reissner’s and Volkersen’s theories) an adhesively bonded joint strength increased with the increase of adhesive thickness [1], however experimentally, most literature reported that the joint strength decreased with the increase of adhesive thickness [2–8]. To obtain a high joint strength, the adhesive thickness should be thin enough, in the range of 0.1 – 0.5 mm [9]; however, it depended on the adhesive type. For very ductile adhesive, such as polyurethane, the adhesive thickness between 0.5 mm and 1 mm gave the optimum strength for aluminium joints [4]. For many structural adhesives, the optimum adhesive thickness was in the range of 0.1 – 0.2 mm [9].

Water can enter into the adhesive joint through several mechanisms; diffusing into the bulk adhesive, wicking into the interface, capillary action into micro voids and cracks, and diffusing into permeable substrate before reaching the adhesive layer [10]. The absorbed water then degrades the mechanical properties of adhesive and affects the interface of adhesive/substrate. Often during aging,
the joint fails prematurely at the interface and is difficult to predict. For simplification (as many factors influences the durability), most of durability studies for metal-metal joints, composite-composite joints and metal composites joints used a constant adhesive thickness assuming the homogeneous water diffusion across the adhesive thickness. In fact, from literature, the optimum adhesive thickness can vary depending on the adhesive type. In moist environment, especially for metal-composite joints, water can diffuse from the adhesive layer and the permeable composites substrate. This can add the complexity of the effect of water on the joint strength. This paper elucidates the effect of water on the steel-glass fiber reinforced unsaturated polyester joints immersed in distilled water at temperature of 60°C at various adhesive thicknesses. Finite element modeling had been undertaken to investigate the water distribution in the adhesive layer as well as in the composites substrate.

2. Experimental methods

2.1. Materials

Low carbon steel was used for substrate with a thickness of 3.96 mm and a width of 30 mm. A room temperature cured Araldite® epoxy was used for adhesive. A composite plate was manufactured by hand lay up using Yukalac BQTN 137 unsaturated polyester resin as matrix and plain weaves glass fiber with an area density of 600 g/m² as the reinforcement. Methyl ethyl ketone peroxide (MEKPO) was used as curing agent (hardener), with resin to hardener ratio of 100:1 (by weight). The thickness of composite plate was 4 mm. The composite was cured at room temperature, under pressure approximately of 0.1 MPa. This composite was then called as glass fiber-reinforced unsaturated polyester (GFRP).

![Figure 1. A single lap joint specimen (not to scale).](image)

2.2. Single lap joint manufacturing

Single lap joints (SLJs) were manufactured using low alloy carbon steel and GFRP substrates, and an Araldite adhesive. The dimension of SLJ is shown in figure 1. Before being bonded, the steel surfaces were abraded using a steel brush and an emery paper (grid 80) to remove the smudge and the contaminants, such as grease and oil. After abrading, they were rinsed using detergent and acetone. The surfaces were subsequently etched using sulphuric acid (30% by weight) for 5 min, cleaned using acetone and wiped using a cloth. For GFRP materials, their surfaces were lightly abraded using an 80 grid emery paper and were cleaned using acetone. The substrates were bonded immediately after the surface treatment using Araldite adhesive. The ratio of resin to hardener was 1:1 (by volume). The thickness of adhesive layer ($t_a$) was varied; 0.1, 0.2, 0.3, 0.4 and 0.5 mm and was maintained by
inserting wires in the overlap area, close to both edges of the overlap. Paper clamps were used to hold the joints during curing. The curing was conducted at a temperature of 120°C for 2 hours. The specimens were conditioned in two conditions; dry and wet conditions. In dry condition, the specimens were tested a day after the adhesive was cured. In wet condition, the specimens were immersed in distilled water at a temperature of 60°C for 15 days (wet) before being tested. A container with a temperature controller was used for immersion.

2.3. Water absorption and the water dependent mechanical properties of adhesive and composites

Water absorption was conducted on the adhesive and the GFRP plate. The water uptake and its water-dependent mechanical properties for Araldite adhesive immersed in the same environment have been conducted in previous research [11]. Water uptake into a GFRP plate was carried out using a plate specimen with dimension of (45 × 45 × 3.25) mm³. The specimens were immersed in distilled water at temperature of 60°C, and periodically taken for weighing. The experimental water uptake (M_t) data were plotted vs. square root of time. Fickian diffusion model, as in equation (1) [12] were fitted with the experimental water uptake data. The diffusion coefficient (D) was obtained from the experimental water uptake data at the linear part of the curve up to M_t/M_∞ of 0.6 using equation (2).

\[
\frac{M_t}{M_\infty} = 1 - \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} \exp \left[ - \frac{(2n+1)^2 \pi^2}{h^2} Dt \right]
\]

\[
D = \frac{\pi}{16} \left( \frac{m \times h}{M_\infty} \right)^2
\]

where \( m \) is the slope of the linear part of water uptake vs. root square time curve, \( t \) is the time, \( h \) is the specimen thickness and \( M_\infty \) is the water uptake at equilibrium.

The tensile specimens of composites were immersed in the same environment as for water uptake specimen. After 15 days, the specimens were taken out from the immersion and immediately tested using a Control universal testing machine, with a load cell capacity of 2000 kN.

3. Experimental results and discussion

3.1. Water uptake and water dependent mechanical properties

Figure 2 shows the water uptake of Araldite adhesive and GFRP with square root of time, while the diffusion parameters are shown in table 1. Fickian diffusion model [12] was plotted along with the experimental water uptakes. For GFRP, in the linear part, the experimental and the Fickian model were in good agreement, but after 3 days immersion (at water uptake of 1.4%), the water uptake started to slow down, deviating from linearity and the Fickian fit curve. The equilibrium water uptake was reached after immersion for about 15 days for both the adhesive and the GFRP. It was observed that water diffusion into the GFRP did not fully follow the Fickian diffusion model. The saturation level of composite was lower (about 55%) than that of Araldite adhesive, but the diffusion constant was almost tenfold higher. For heterogeneous materials such as GFRP, the diffusion rate can be faster as the water molecules can wick into the interface of matrix and glass fiber, in addition to the diffusion from the matrix [10].
After 15 days immersion, the GFRP specimens were tested under tension and the mechanical properties are shown in Table 2. Referring to Figure 2, after 15 days immersion, the composite has reached the saturation level. In this condition, as seen in Table 2, the decrease of elastic modulus and the tensile strength seemed not to be significant as the standard deviation was high. For woven glass fiber-reinforced polymer composites, the tensile strength ($X$) and the elastic modulus ($E$) in longitudinal (indicated by 11 subscript) and transverse (indicated by 22 subscript) directions were the same. The adhesive properties in dry and wet (after 15 days immersion) conditions obtained from published literature [11], can be seen in Table 3. The adhesive tensile properties degraded considerably after ageing in distilled water at a temperature of 60°C for 15 days. The decrease of elastic modulus and tensile strength was 83% and 71%, respectively.

**Figure 2.** Water uptakes of GFRP and adhesive.

**Table 1.** The diffusion parameters of adhesive and GFRP.

| Materials  | $M_\infty$ (%) | $D$ (mm$^2$/day) |
|------------|---------------|------------------|
| Adhesive   | 4.15          | 0.072            |
| GFRP       | 1.88          | 0.61             |

**Table 2.** The GFRP properties in dry and wet conditions.

| Condition | GFRP |
|-----------|------|
|           | $E_{11} = E_{22}$ (GPa) | $X_{11} = X_{22}$ (MPa) |
| Dry       | 12.39 ± 1.69 | 301.15 ± 21.2 |
| Wet       | 11.00 ± 0.64 | 296.78 ± 5.24 |

**Table 3.** The adhesive properties in dry and wet conditions.

| Condition | Araldite adhesive [11] |
|-----------|------------------------|
|           | $E$ (MPa) | Tensile strength (MPa) |
| Dry       | 2500    | 41.29 ± 4.65 |
| Wet       | 420     | 12.06 ± 3.10 |
3.2. Residual static strength of aged single lap joint

Figure 3(a) shows the shear strength of steel-GFRP composites joints in dry and wet conditions at various adhesive thicknesses. It is seen that in dry condition, the shear strength tended to increase linearly with the increase of adhesive thickness up to 0.5 mm. At the thickness from 0.2 to 0.5 mm, the variation, shown by the vertical bars in the graph, was high. It might be caused by the different mode of failure of each joint, either interfacial or mix failure. Sugiman et al. [11] also reported the same trend on the shear strength of bonded steel joints with the adhesive thickness using the same adhesive as used in this study. Generally, the shear strength decreased with the increase of adhesive thickness [2–8], as the increase of adhesive thickness tended to have more flaws [1]. The increase of shear strength in this case, may be caused by the increase of plastic deformation in the fracture zone as the adhesive is quite ductile. Campilho et al. [13] found that the critical energy release rate of joints with a ductile polyurethane adhesive increased essentially linear with the increase of adhesive thickness up to 2 mm. This was due to enlargement of adhesive plastic zone in front of crack tip in the joint with the increase of adhesive thickness.

Figure 3. Showing, (A) the shear strength with adhesive thickness and (B) the failure surfaces of steel-GFRP bonded joints, in dry and wet conditions. In the right figure, symbols a, b, c, d, and e are respectively showing the adhesive thickness of 0.1 mm, 0.2 mm, 0.3 mm, 0.4 mm and 0.5 mm.

In wet condition, the joint strength as seen in figure 3(a), was very low and seemed slightly increasing with the adhesive thickness. This could be due to the absorbed water that influenced the adhesive bulk strength and attacked the interface adhesive/substrate. Interestingly, at the adhesive thickness of 0.1 mm, the dry and the wet joint strengths were about the same, while the strength of other thicknesses in wet condition dropped considerably compared to the dry joints strength. This is interesting as in our previous study [11], in wet condition, on the steel-steel joints, the joint strength still increased with the increase of adhesive thickness up to 0.3 mm, and then dropped steeply afterward.

As seen in figure 3(b), in dry condition, mode of failure changed from interfacial (at 0.1 mm) to mix failure (0.2 mm - 0.5 mm) with the increase of adhesive thickness. The mix failure was indicated by the tearing of adhesive and by the delamination of part of the GFRP, adhering to the steel substrate. The extent of mix failure increased as the size of the delaminated GFRP got bigger and bigger with the increase of adhesive thickness. It is expected as the mix failure gives higher joint strength than the interfacial failure. However, the cause of this kind of failure needs to be further investigated by analysing the stress state of the joints.
In wet condition, modes of failure were mostly interfacial at the adhesive-steel interface, regardless the adhesive thickness. The interfacial failure occurred at the adhesive-steel interface rather than at the adhesive-GFRP interface, because the adhesive-metal interface was not stable in water environment [14, 15]. Moreover, the steel tended to corrode after being aged in water. The corrosion product might further displace the adhesive from the steel surface, decreasing the joints strength [16]. Some corrosion products were seen at the steel surface after failure. After 15 days immersion, the adhesive and the adhesive-steel and the adhesive-GFRP interfaces had been wetted by water, even though the water content at both interfaces was different. This will be discussed further in the water diffusion modelling section.

4. Finite element modelling of moisture diffusion

To obtain water distribution in the adhesive layer and in the composites substrate, a finite element modelling was carried out using ABAQUS utilizing a mass diffusion analysis. The detail of this method can be found elsewhere in ABAQUS documentation [17]. For Fick’s law, the concentration flux, $F$ can be described as in equation (3),

$$F = -sD \left( \frac{\partial \phi}{\partial x} \right)$$

The normalized concentration ($\phi$) along the diffusion path $x$, is defined as $c/S$, where $c$ is the mass concentration and $S$ is the solubility of diffusing substance in base material. Two parameters are required to model the water diffusion using mass diffusion; diffusivity ($D$) and solubility ($S$), corresponding to the diffusion rate and the equilibrium water uptake, respectively.

Finite element modelling was performed in 3-dimensional (3D) with meshing as seen in figure 4. There were four elements across the adhesive thickness, so that the thickness of an element was 0.025, 0.05, 0.075, 0.08, 0.125 mm for the adhesive thickness of 0.1, 0.2, 0.3, 0.4 and 0.5 mm, respectively. Both of the element length and width were 0.5 mm.

**Figure 4.** Meshing of finite element modelling for water diffusion in adhesive and GFRP.

The diffusion parameters of GFRP and the adhesive discussed in the previous section were used as the input parameters in the diffusion finite element modelling. Water content was normalised using the equilibrium water uptake and then it was called as the normalised concentration (NNC). Water diffused from four sides of the adhesive layer of the joint and from the thickness direction of the GFRP. Water diffusion from the GFRP width sides was not modelled because in practice water usually came in from the thickness direction.

Figure 5 shows the contour plot of water distribution in the GFRP and the adhesive layer at the various adhesive thicknesses, while the distribution of water both in the adhesive layer along the
overlap length and in the through thickness of the joint is shown in figure 6. The contour shows a half of width wise of the overlap region and the water distribution plots shown in figure 6 were taken at the midplane of the overlap length. As seen in figure 6(a), water distribution across the adhesive thickness shown by the normalised concentration, seemed not homogeneous and it was more obvious with the increase of adhesive thickness. The water diffusion from four sides of adhesive layer was not different in all adhesive thicknesses; however, the water diffusion from the through thickness of GFRP across the adhesive layer contributed to the difference of water content at the adhesive-steel and adhesive-GFRP interfaces. As water came in first from the GFRP, the water content at the adhesive-GFRP interface was higher than at the adhesive-steel interface. At the thickness of 0.1 mm, water content was more homogeneous, but as the thickness increased, the different of water content at both interfaces was more pronounced. After 15 days immersion, the saturation in the adhesive layer had not been reached yet. At the middle of the joint, the normalised concentration was around 0.7 and 0.5 for the adhesive thickness of 0.1 and 0.5 mm, respectively.

In figure 6(b), although the immersion time was the same, water distribution across the GFRP thickness and the adhesive layer thickness was not the same. At the adhesive thickness of 0.1 mm, the water content both in the GFRP and the adhesive layer was the highest and more homogeneous across the adhesive layer; however, as the adhesive thickness increased, more water diffused from GFRP into the adhesive layer, so that at the same frame time, water distribution in GFRP and in adhesive layer shifted to the lower value. The decrease of water content from the adhesive thickness of 0.1 mm to 0.5 mm was about 36%. As the water content at the adhesive thickness of 0.5 mm was lower than that in 0.1 mm, the joint strength at the adhesive thickness of 0.5 mm should be higher. However, the experimental results showed that the joint strength at all the adhesive thicknesses seemed not significantly different. It suggests that there is critical water content at the adhesive-steel interface above which the joint strength tends to be constant. Loh et al. [18] reported that the joint strength
decreased sharply up to the water content of 3% and tended to level off afterward. Referring to figure 6(b), the water content was higher than 3%, so that it is consistent with that in [18].

![Diagram showing distribution of water across adhesive layer thickness along overlap length](image)

**Figure 6.** Showing distribution of (a) water across the adhesive layer thickness along the overlap length, (b) water in the GFRP and adhesive layer. The distribution in (a) was taken at the middle of width wise, while the distribution in (b) was taken at the center point of the joint.

5. Conclusions

Experimental studies on water diffusion in the glass fiber reinforced unsaturated polyester (GFRP) and in the Araldite adhesive and its effect on the tensile properties had been undertaken. The investigation of the effect of adhesive thickness on the joints strength had also been carried out after being immersed in distilled water at a temperature of 60°C for 15 days. It was found that the equilibrium water content of the GFRP was lower than that of the Araldite adhesive, but the diffusion rate was much higher. The effect of water on the tensile properties of GFRP seemed not to be significant, whereas its effect on the tensile properties of adhesive was significant. In dry condition, the joint strength tended to increase with the increase of adhesive thickness; however, in wet condition, the effect of adhesive thickness appeared not to be significant. At the thickness of 0.1 mm, the joint strength in dry and in wet conditions was about the same, but the effect of water was more obvious at the thicker adhesive thickness. Finite element modelling on water diffusion had been undertaken utilizing a mass diffusion analysis. Water content across the adhesive thickness was not homogeneous and it was more noticeable at the thicker adhesive layer. The water content at the adhesive-GFRP interface was higher than at the adhesive-steel interface due to the contribution of water from the GFRP.

Acknowledgements

This work was supported by Ministry of Research, Technology, and Higher Education, the Republic of Indonesia with grant number [733/UN18.L1/PP/2018]. Thanks also to I Wayan Januartha for specimens preparation.

References

[1] Adams R D and Peppiatt N A 1974 Stress analysis of adhesive-bonded lap joints *Journal of Strain Analysis* **9** 185–96

[2] Yang S, Xu W, Liang L, Wang T and Wei Y 2014 An experimental study on the dependence of the strength of adhesively bonded joints with thickness and mechanical properties of the adhesives *Journal of Adhesion Science and Technology* **28**(11) 1055–71
[3] da Silva L F M, Rodrigues T N S S, Figueiredo M A V, de Moura M F S F and Chousal J A G 2006 Effect of adhesive type and thickness on the lap shear strength The Journal of Adhesion 82(11) 1091–1115

[4] Boutar Y, Naími S, Mezlini S, da Silva L F M, Hamdaoui M and Ali M B S 2016 Effect of adhesive thickness and surface roughness on the shear strength of aluminium one-component polyurethane adhesive single-lap joints for automotive applications Journal of Adhesion Science and Technology 30(17) 1913–29

[5] Adnan A and Sun C T 2008 Effect of adhesive thickness on joint strength: A molecular dynamics perspective The Journal of Adhesion 84(5) 401–20

[6] Castagnetti D, Spaggiari A and Dragoni E 2011 Effect of bondline thickness on the static strength of structural adhesives under nearly-homogeneous shear stresses The Journal of Adhesion 87(7–8) 780–803

[7] van Tooren M J L, Gleich D M and Beukers A 2004 Experimental verification of a stress singularity model to predict the effect of bondline thickness on joint strength Journal of Adhesion Science and Technology 18(4) 395–412

[8] Kahraman R, Sunar M and Yilbas B 2008 Influence of adhesive thickness and filler content on the mechanical performance of aluminum single-lap joints bonded with aluminum powder filled epoxy adhesive Journal of Materials Processing Technology 205 183–9

[9] Gleich D M, Van Tooren M J L and Beukers A 2001 Analysis and evaluation of bondline thickness effects on failure load in adhesively bonded structures Journal of Adhesion Science and Technology 15(9) 1091–1101

[10] Adams R D, Cowap J W, Farquharson G, Margary G M and Vaughn D 2009 The relative merits of the Boeing wedge test and double cantilever beam test for assessing the durability of adhesively bonded joints, with particular reference to the use of fracture mechanics International Journal of Adhesion and Adhesives 29 609–20

[11] Sugiman, Akbar I, Sulistyowati E D and Setyawan P D 2016 Effect of adhesive layer thickness on the shear strength of adhesively bonded steel joints in wet environment Applied Mechanics and Materials 836 78–82

[12] Crank J 1975 The mathematic of diffusion, 2nd ed. (London: Oxford University Press)

[13] Campilho R D S G, Moura D C, Banea M D and da Silva L F M 2014 Adhesive thickness effects of a ductile adhesive by optical measurement techniques International Journal of Adhesion and Adhesives 57 125–32

[14] Kinloch A J, Dukes W A and Gledhill R A 1975 Durability in adhesive joints, 2 Polymer Science and Technology vol 9B, ed L H Lee (New York: Plenum Press) pp 597–614

[15] Kinloch A J 1979 Interfacial fracture mechanical aspects of adhesive bonded joints – A review The Journal of Adhesion 10(3) 193 – 219

[16] Zanni-Deffarges M P and Shanahan M E R 1995 Diffusion of water into an epoxy adhesive: comparison between bulk behaviour and adhesive joints International Journal of Adhesion and Adhesives 15 137–42

[17] ABAQUS 2009 User manual version 6.9.1 (USA: Habit, Karlsson & Sorensen, Inc.)

[18] Loh W K, Crocombe A D, Wahab M M A and Aschcroft I A 2002 Environmental degradation of the interfacial fracture energy in adhesively bonded joint Engineering Fracture Mechanics 69 2113–28