Investigation of technological factors influencing the strength of bonded Al – alloy

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Abstract. Adhesive bonding makes up a significant part of rigid joints of machine elements. The aim of this study was to investigate the contamination effect of moisture and oil on the bond strength of aluminium alloy 2024 T3 bonded by film and liquid adhesives using mechanical and chemical surface preparation methods. Two types of joints were used: homogeneous (Al-Al) and heterogeneous (Al-GFRP) ones. Surface preparation highly increased surface roughness Ra. The surfaces were moistened with deionized water and mineral oil and glued after a constrained time delay (outlife). Tensile shear strength tests of the joints showed higher strength of the chemically prepared surfaces by gluing with film adhesive. Contrarily mechanically prepared surfaces were stronger with liquid adhesive. Film adhesive seemed less sensitive to surface contamination in general.

1. Introduction
Aluminium alloys are one of the most important construction materials [1]. The global aluminium consumption volume of nearly 68.6 metric tones attained in 2019 and it is projected to reach almost 85 mln. metric tons by 2025 [2]. Aluminium alloys have wide industrial use (automotive, aviation etc.). Lighter vehicles lower fuel consumption (reduce CO₂ emissions) and more than 90% recyclability makes aluminium an ideal material for circular economy [1].

Adhesive bonding of aluminium alloys started in 1940’s in England and since then has become more popular among other traditional connections (riveting, welding, etc) [3]. Bonding is simple and effective type of connection not reducing the strength of constructional elements [4]. Adhesive bonding is the most suitable method of joining both for metallic and non–metallic structures where strength, stiffness and fatigue life must be maximized at a minimum weight [5]. Mechanical interaction and degradation of bonds are complex, strength of the connection is influenced by various factors – materials used, geometrical parameters, etc. Degradation in a bonded joint begins with a mixed load, both the load and the operating environment must be considered, as the joint is affected by temperature, humidity, lubricants, etc. [6].

Surface preparation is particularly important for the joint strength and durability. Its influence on surface preparation of glued joints was investigated [7]. Aluminium is special in that its surface is coated with an oxide layer, so various surface preparation methods are used the most effective of which are combined ones: degreasing, etching, anodizing, etc. [8, 9]. The strength of the connection in operation is influenced by the environment, the most important factor of which is humidity. Water enters through
surfaces, cracks and other defects in adhesive and adherend surfaces, weakening the bond and accelerating loss of adhesion [10].

The aim of the research is to investigate the influence of technological factors on the strength of the bonded aluminium alloy joint by evaluating the influence of surface contamination with water and mineral oil.

2. Materials and methods

2.1. Materials

There were 126 specimens prepared for testing all of the same dimensions in length and width being 60x15 mm (gluing area 15x15=225 mm²) and 6×3 test plan was selected with 6 groups of materials and 3 subgroups in each. The plan is presented in table 1.

| Code | Adher. | Adhes. | Surface prepar. | Surface contamination | Constrained time delay | Strength of 126 samples | Elongation at break of 126 |
|------|--------|--------|----------------|-----------------------|------------------------|-------------------------|---------------------------|
| 1AK  | Al-Al  | film   | mech           | uncontaminated         | 0                      | n=3                     | n=3                       |
| 1A1  | Al-Al  | film   | mech           | water                 | 5, 15, 30 min          | n=9(3x3)                | n=9                       |
| 1A2  | Al-Al  | film   | mech           | min. oil              | 1, 12, 24 h            | n=9                     | n=9                       |
| 2AK  | Al-Al  | film   | chem           | uncontaminated         | 0                      | n=3                     | n=3                       |
| 2A1  | Al-Al  | film   | chem           | water                 | 5, 15, 30 min          | n=9                     | n=9                       |
| 2A2  | Al-Al  | film   | chem           | min. oil              | 1, 12, 24 h            | n=9                     | n=9                       |
| 3AK  | Al-Al  | liquid | mech           | uncontaminated         | 0                      | n=3                     | n=3                       |
| 3A1  | Al-Al  | liquid | mech           | water                 | 5, 15, 30 min          | n=9(3x3)                | n=9                       |
| 3A2  | Al-Al  | liquid | mech           | min. oil              | 1, 12, 24 h            | n=9                     | n=9                       |
| 4AK  | Al-Al  | liquid | chem           | uncontaminated         | 0                      | n=3                     | n=3                       |
| 4A1  | Al-Al  | liquid | chem           | water                 | 5, 15, 30 min          | n=9                     | n=9                       |
| 4A2  | Al-Al  | liquid | chem           | min. oil              | 1, 12, 24 h            | n=9                     | n=9                       |
| 5AGK | Al-GFRP| liquid | mech           | uncontaminated         | 0                      | n=3                     | n=3                       |
| 5A1  | Al-GFRP| liquid | mech           | water                 | 5, 15, 30 min          | n=9(3x3)                | n=9                       |
| 5A2  | Al-GFRP| liquid | mech           | min. oil              | 1, 12, 24 h            | n=9                     | n=9                       |
| 6AGK | Al-GFRP| film   | mech           | uncontaminated         | 0                      | n=3                     | n=3                       |
| 6A1  | Al-GFRP| film   | mech           | water                 | 5, 15, 30 min          | n=9                     | n=9                       |
| 6A2  | Al-GFRP| film   | mech           | min. oil              | 1, 12, 24 h            | n=9                     | n=9                       |

Total: n=126 n=126

4 groups consisted of homogeneous adherends (Al-Al), the remaining groups consisted of heterogeneous (Al – GFRP) ones. Two adherend materials were used: aluminium alloy 2024 T3 and glass fiber Gilliner 1076C Type59 (E – glass, polyester resin system). Aluminium alloy chemical composition is presented in table 2. The mechanical properties of fiberglass are given in table 3. The properties in the warp and weft directions are calculated and given averaged.

There were 3 subgroups in each group: uncontaminated (control joints) and contaminated (with deionized water and mineral oil). Low viscosity mineral oil AeroShell Fluid3 was used.

Epoxy adhesive types used: film AF163–2K and liquid EC 2216 B/A Gray (two component). Both are 3M (USA) materials. The technical data of the adhesives are given in table 4. Thickness of dried film epoxy adhesive is 0.25 mm, liquid – 0.1 mm, GFRP – 1.45 mm.

| Code | Adher. | Adhes. | Surface prepar. | Surface contamination | Constrained time delay | Strength of 126 samples | Elongation at break of 126 |
|------|--------|--------|----------------|-----------------------|------------------------|-------------------------|---------------------------|

Table 1. Design of experiment (DOE) matrix

Table 2. Material properties of Al 2024-T3 alloy [11]

| Main chemical composition Al 2024-T3 alloy [wt. %] |
|---------------------------------------------------|
| Al | Cu | Mg | Mn | Fe | Zn | Ti | Cr |
| 90.7-94.7 | 3.8-4.9 | 1.2-1.8 | 0.3-0.9 | ≤ 0.5 | ≤ 0.25 | ≤ 0.15 | ≤ 0.1 |
2. Surface preparation and testing

Before gluing surfaces were prepared mechanically (mech) and chemically (chem). During mechanical preparation surfaces were abraded with P180 emery paper in 3 angular directions (90°, 180°, 45°), cleaned with acetone and after water film test coated with the chromate conversion coating Henkel BODERITE M–CR 1200S RTU AERO. During chemical preparation, surfaces were cleaned with NaOH solution and afterwards kept in the sulfuric acid– dichromate (FPL (Forest Product Laboratory) is a long used surface treatment for Al in the aerospace industry) solution for 60 s. Surface roughness in µm Ra was measured with MarSurf PS10 (Mahr, Germany) device. Gluing was done with a special gluing station HC9200B (HEATCON, USA), connections heated and joined by vacuum (0.88 bar). The samples with film and liquid adhesives were heated and cooled at the same rate of 16.1°C/min and all were cooled down till 25°C. The samples with film adhesive were heated till 121.1°C and hold for 90 min, while with liquid adhesive were heated till 93.3°C and hold for 120 min.

Surface contamination was created by shortly immersing planned part of samples to be bonded into water, the other ones – into oil. The test samples were kept in a special holder to remove excess of liquid and then after a set time were bonded at 22±1°C temperature and 40±5% humidity. Samples taken from water were bonded after 5, 15 and 30 min, from oil – after 1, 12 and 24 hours. Tensile shear strength tests made with INSTRON 5965, tension speed 20 mm/min, one end of the samples were immovable. Recorded were max force and elongation at break according to ISO 4587 standard [13], calculations done by the equation: \( \tau = \frac{F_{\text{max}}}{LW} \). Here \( \tau \) – tensile shear strenght [MPa], \( F_{\text{max}} \) – maximum force [N]; \( L \) – lapping length [mm]; \( W \) – lapping width [mm].

The fracture structure was described according to EN ISO 10365:1995 standard [14]: SF (substrate failure), CSF (cohesive substrate failure), DF– delamination failure, CF (cohesive failure), SCF (special cohesion failure), AF (adhesion failure), ACFP (adhesion and cohesion failure with peel). The number next to the letter indicates the amount of disintegration as a percentage.

Software packages IBM SPSS Statistics 2020, MS Excel add-in Data Analysis and add-on XLSTAT were used for data entry and analysis.

3. Experimental results and discussion

Surface roughness of the aluminium alloy 2024 T3 before preparation was \( R_a = 0.22 \pm 0.032 \) (the standard error SE= 0.018); ground in 3 directions – 0.95 ±0.080 (SE= 0.046); ground and covered with the...
conversion coating – 0.93 ±0.049 (SE=0.028); cleaned with NaOH solution – 0.83 ±0.075 (SE= 0.043); etched in the FPL acid solution – 0.85 ±0.027 (SE= 0.015).

Close to one $\eta=0.986$ value received after Crosstabs analysis show high degree of association between variables (surface preparation and roughness). Generalized linear model allows to state that 97.2% of the scatter of roughness values are influenced by the surface preparation methods used (the coef. of determ. $R^2=0.972$, $p=0.05$).

Based on the tensile and elongation at break test data mathematical bond failure model was developed and 50% failure probability estimated, taking tensile and elongation at break separately as failure criteria (Figure 1). Sample size of each group n=21 (minimum bound ratio requirements of n=20 are being met for this Weibull analysis).

![Figure 1. Weibull plot (estimation of parameters by using log-linear regression)](image)

Data are presented in table 5. Higher values of Weibull modulus m are observed of the strongest bonds (it shows lower standard deviation and lower spread of failure criteria values from the mean).

| Bond failure criterion – elongation at break: |  |
|---|---|
| Group | Bond description | Regression equation $y=15.28x$ - 4.11, $R^2=0.962$ |
| 1AK-1A1-1A2 | Al-Al mech-film | 1.28 15.28 1.31 |
| 2AK-2A1-2A2 | Al-Al chem-film | 1.76 15.20 1.81 |
| 3AK-3A1-3A2 | Al-Al mech-liquid | 1.02 9.18 1.06 |
| 4AK-4A1-4A2 | Al-Al chem-liquid | 0.91 4.34 1.01 |
| 5AK-5A1-5A2 | Al-GFRP mech-liq. | 0.95 7.91 1.00 |
| 6AK-6A1-6A2 | Al-GFRP mech-film | 0.96 3.96 1.06 |

| Bond failure criterion – shear tensile strength: | $\tau$, [MPa] |
|---|---|
| 1AK-1A1-1A2 | Al-Al mech-film | $y=9.99x$-28.64, $R^2=0.987$ |
| 2AK-2A1-2A2 | Al-Al chem-film | $y=29.59x$-92.16, $R^2=0.987$ |
| 3AK-3A1-3A2 | Al-Al mech-liquid | $y=7.58x$-19.91, $R^2=0.998$ |
| 4AK-4A1-4A2 | Al-Al chem-liquid | $y=6.83x$-16.86, $R^2=0.946$ |
| 5AK-5A1-5A2 | Al-GFRP mech-liq. | $y=5.30x$-12.49, $R^2=0.979$ |
| 6AK-6A1-6A2 | Al-GFRP mech-film | $y=6.74x$-16.55, $R^2=0.977$ |

Figure 2 shows contaminated bonds strength with varied time between contamination and bonding. An average strength of uncontaminated surfaces is given at the 0 time or before the test. For statistical evaluation of results XLSTAT mixed linear model was used with 6 groups of materials and 7 constrained time delay intervals (a–0 min (control), b–c–d (5–15–30 min H$_2$O) and e–f–g (1–12–24 h min. oil) were selected and bond strength evaluated.
Figure 2. Impact of time delay on the avg. change of strength of contaminated adhesive bonds with water (a) and oil (b)

It has been determined that time delay interval after contamination has a great influence on the model (error covariance $p<0.0001$) and doesn’t have on covariance among material groups, but is on the limit ($p=0.058$). Statistically the most significant are uncontaminated samples ($p<0.0001$) when compared with contaminated ones and time interval until the first measurements of the strength of the contaminated bonds (the less time elapsed since the start of contamination, the weakest are the bonds).

Time delay after contamination with water is insignificant from the period of 5 min ($p=0.493<0.05$) till 15 min ($p=0.191<0.05$), slow change of strength is observed, however, after 30 min elapsed it started to show higher impact ($p=0.006$). Time delay after contamination with oil has the highest impact on strength after 1 ($p<0.001$) and 12 ($p<0.009$) hours elapsed, significant change of strength is observed.

Statistically significant are only two groups: 2nd ($p=0.21\times10^{-3}$) – contamination with water or oil has the lowest influence on the average strength and was the highest in average strength values, 5th ($p=0.04$) – the lowest average strength (table 6).

| Contamination | Mechanical treatment | Chemical treatment | Avg. |
|---------------|----------------------|--------------------|------|
| Water         |                      |                    |      |
| Uncont. ($t_0$) avg. strength, [MPa] | 1A1 film | 2A1 film | 3A1 liquid | 5A1 liquid | 6A1 film | Avg. |
| Reduction of avg. strength [MPa] ($t_0$–$t_{30}$ min) | 18.58 | 15.21 | 12.94 | 13.12 | 22.75 | 12.98 | 15.93 |
| Fracture denomination of contaminated bond: | AF100 | AF100 | DF10 | AF90 | DF80 | AF20 | CF100 | AF100 start, AF30CF70 end |
| Mineral oil   |                      |                    |      |
| Uncont. ($t_0$) avg. strength, [MPa] | 1A2 film | 2A2 film | 3A2 liquid | 5A2 liquid | 6A2 film | Avg. |
| Reduction of avg. strength [MPa] ($t_0$–$t_{43}$ min) | 18.58 | 15.21 | 12.94 | 13.12 | 22.75 | 12.98 | 15.93 |
| Fracture denomination of contaminated bond: | AF100 | AF100 | DF85 | AF70 | AF25 | CF75 | AF100 |
Images of the strongest and weakest disintegrated adhesive bonds are presented in figure 3.

![Images of fracture surfaces](image)

**Figure 3.** Images of fracture surfaces: a) the strongest Al – Al (group 2), the weakest groups: b) Al – GFRP (group 5) and c) Al – GFRP (group 6)

As can be seen from the image (a), the strongest bonds failed in cohesion. These are chemically prepared surfaces and thermal gluing of film adhesive in vacuum greatly increased adhesion. In general film adhesive is nearly twice higher in technological thickness compared to liquid ones and have get better mechanical properties. Meanwhile the weakest bonds are those with dissimilar materials (Al-GFRP) which surfaces were prepared mechanically. Despite they are liquid (b) or film (c) ones, it can be explicitly seen, that they mainly failed due to delamination of fibers with the most severe damages and delamination observed on the glass laminate surfaces. It reveals weaker interlaminar strength of the glass fiber in tensile shear. In order to increase the bond strength between metal and glass fiber, stronger then polyester fibers and resins should be used, special structural primer applied, etc. Besides, it is know that in case of differences in the thickness and type of adherend materials, due to eccentricity such single lap bonds are impacted by higher forces and moments and their failure seems more complex compared to similar ones [6,7].

4. Conclusions

- Surface roughness and scatter of its values is 97.2% affected by the surface preparation method used. The most uniform distribution of roughness Ra values achieved with FPL etch solution—the lowest standard deviation and error (SD = ±0.027, SE= 0.015).
- Contamination with water is less dangerous than with mineral oil (strength recovers faster). Bonds contaminated with water have lower strength by 5% in average, type of the adhesive does not influence the strength of the bond. The lowest strength decrease has been observed of bonds with homogeneous materials (Al-Al) and both by chemical (0.02%) and mechanical (2.2%) surface preparation methods.
- Contamination with oil reduces the strength by 14% in average. The hybrid material (Al-GFRP) bonds mechanically prepared and glued with liquid glue has the highest reduction of strength (~24%).
- Mechanically prepared AL-GFRP and contaminated with water and oil bonds showed reduction in strength in 10-20% (film-liquid), prevailing mixed disintegration DF+AF.

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