The effect of laser surface treatment on the adhesive bonding performance of aluminum alloy

W Wang, J W Xu, X Chen, Y J Liang and L Z Zhu*
Nanjing Research Institute of Electronics Technology, Nanjing 210039, China
druid_kurama@sina.com

Abstract. For the purpose of convenient treatment before gluing and improving the adhesive bonding performance, we compared the tensile shear strengths of epoxy adhesive bonding to 2A12 aluminum with preferred pre-treatments. Taking advantages on both convenience and pre-treatment quality, we investigated the influences of duration of laser treatment on the adhesive bonding strength to obtain the optimized extents of laser treatment. With the analysis of scanning electron microscopy, laser confocal scanning microscopy and contact angle measurements adopted to characterize the surface morphology, roughness and wettability, respectively, we discussed the effects of pre-treatments on the surface status and adhesive bonding. As a result, we proposed a pre-treatment with laser treatment on aluminum to improve adhesive bonding performance, by which the removal of oxide layer, surface roughening and cleaning were easily accomplished before gluing.

1. Introduction

Aluminum alloys have been widely used in industrial fields with great demands on lightweight structures such as automobile, electronic equipment and aerospace, because of their excellent mechanical property, good processability and relatively low density. The aluminum structures to themselves or the structures with other kind of materials are often assembled together by connecting with mechanical fastening, welding and gluing. Taking advantages of limited increase in weight, low cost, good applicability and well distributed stresses, the gluing method has been extensively applied in the connecting of aluminum structures [1-5].

The mechanical performance of adhesive joint of aluminum to aluminum or other composite highly depends on the bonding strength between the adhesive and the substrate. Appropriate pre-treatment can not only remove all contaminants on the surface, but also modify the surface conditions such as improving the roughness, wettability, surface energy, and introducing functional groups in the surface, which significantly enhances the mechanical properties of the bond [4-9]. Traditional methods for pre-treatments include mechanical, chemical and electrochemical treatments, the most common are mechanical roughening, etching and anodizing [6]. However, adhesive joints with mechanical roughening samples present low strength and durability and risk contamination of the surface during the process. As for chemical and electrochemical treatments, they involve in higher costs than mechanical method and environmental pollution problems.

Laser surface treatment is a dry treatment, which prevents extremely dangerous to health and environment and possible containments to the surface. A focused beam produced by the laser equipment generates well controlled surface treatment with ablating and even peeling off the outmost layer to improve roughness, wettability and surface energy contributing to satisfactory promotion to interface bonding performance, by the mechanism of thermal vibration, vaporization, fusion and
plasma stripping to clean and etch the surface [5-7]. Benefiting from superiorities in high quality cleanliness, precise treated area and convenient and high-efficiency treatments, the laser surface treatment has been used in coating removing, rust removing, industrial mold cleaning and preservation of cultural relics [6-11].

For the purpose of convenient treatment before gluing and improving the adhesive bonding performance, we compared the tensile shear strengths of epoxy adhesive bonding to 2A12 aluminum with preferred pre-treatments. Taking advantages on both convenience and pre-treatment quality, we investigated the influences of duration of laser treatment on the adhesive bonding strength to obtain the optimized extents of laser treatment. With the analysis of scanning electron microscopy (SEM), laser confocal scanning microscopy (LCSM) and contact angle measurements adopted to characterize the surface morphology, roughness and wettability, respectively, we discussed the effects of pre-treatments on the surface status and adhesive bonding. As a result, we proposed a pre-treatment with laser treatment on aluminum to improve adhesive bonding performance, by which the high efficiency removal of oxide layer, surface roughening and high-quality cleaning were easily accomplished before gluing.

2. Experiments

2.1. Samples

According to international standard of ISO 4587: 2003, the samples were prepared by cutting 2A12 aluminum sheets into 100 mm × 25 mm × 2 mm pieces. Considering that chemical conversion treatment was widely used in electronic equipment, such as enclosures, crates and cabinets due to the requirements on surface conductivity, the pieces were treated by chemical conversion method to form low resistance oxide films which followed Chinese standard in electronic field of SJ 20813-2002 in order to correspond to the actual using state. Except for the BLANK samples as cut status for control experiment, others samples were treated by chemical conversion then pre-treatments before gluing.

Table 1. The method of pre-treatment before gluing.

| Pre-treatment    | Detail                                                                 |
|------------------|------------------------------------------------------------------------|
| Grinding         | Pneumatic grinding with 80 or 120 mesh sandpaper until tarnished.      |
| Sand blasting    | Blasting with 80 mesh quartz sand under the pressure of 0.6 MPa.       |
| Laser treatment  | Carrying out the laser treatment by a domestic commercial equipment for 2 to 12 s, which contained the laser generator with nominal power at 50 W, wavelength at 532 nm and pulse repetition rate in the range of 10–600 kHz. The focused laser beam uniformly scanned in speed of 4000 mm/s within the area setting as 30 mm × 35 mm. |

Figure 1. Schematic of bonded aluminum samples in a pair.

According to ISO 4587: 2003, a domestic epoxy adhesive was spread evenly onto the adhesion area to bond together the two pieces of aluminum samples as shown in Figure 1, and the thickness of adhesive was controlled by inserting glass spheres with diameter of 0.2 mm in the bonding area. The adhesive between the two pieces was pressurized for 2 h and then cured for 72 h in room temperature.
2.2. Characterization

2.2.1. Adhesion strength. The adhesion strength was determined from tensile shear test in a universal tensile testing machine according to ISO 4587: 2003. And both ends in bonded samples were stick with gaskets of 2 mm thickness before clamping by the holder, to avoid the measuring errors caused by torque during the tensile shear tests.

2.2.2. Surface morphology. The surface morphology was characterized by Zeiss Gemini-500 SEM. The surface profile and roughness were obtained with Keyence VK-9710 LCSM, and the sampling area for roughness measuring was set as X=1024 μm and Y=768 μm.

2.2.3. Contact angle. The contact angle of water was measured by a Dataphysics OCA-20 optical contact angle meter, and the test was conducted by means of sessile drop technique at room temperature. A drop of 2 μL of water was placed on the surface and the contact angle was determined after 30 s.

3. Results and discussion

3.1. The adhesive bonding performance of aluminum samples with pre-treatments

For the requirements on surface conductivity, the aluminum structures of electronic equipment such as enclosures, crates and cabinets were often treated with chemical conversion, however, the limited mechanical strength of chemical conversion layer made it unsuitable in high-performance adhesive joint [11, 12]. In our study, several pre-treatments of grinding, sand blasting and laser treatment were involved in removing the oxidation layers, and improving the surface roughness which could provide more interface area to bond.

As shown in Figure 2a, the chemical conversion layers were removed by pre-treatments from a macro view, as for laser treatment the removal was achieved with the scanning time above 2 s. The destructions of glued joint of bonded samples with pre-treatments were observed in Figure 2b, which showed that the peeling off phenomenons happened from the interface between metal and adhesive.

![Figure 2](image_url)

Figure 2. The aluminum samples after pre-treatments (a) and the destructions of adhesive joint of bonded samples in pairs (b) (the samples were pre-treated with (1) blank, (2) grinding by 80 mesh sandpaper, (3) sand blasting, laser treatment for (4) 2 s and (5) 9 s).

The adhesive bonding strength with different pre-treatments was shown in Figure 3a, the blank sample appeared an adhesive strength of 4.0 MPa, and the adhesive strengths were significantly increased with pre-treatments. For the samples pre-treated with grinding, the adhesive strength reached from 10.5 to 12.4 MPa, and the sample grinded with coarser sandpaper showed stronger adhesive strength, which could ascribe that the coarser sandpaper employed the rougher surface formed. As sand blasting was a high-quality pre-treatment method, the adhesive strength with it rise to 17.2 MPa. Although the pre-treatment of sand blasting performed best in adhesive bonding performance, which could cause distortion and damage to the work-pieces and give limited applicability to assembling
stage and precision work-pieces. And with laser treatment for 9 s, the adhesive strength was nearly equal to that with sand blasting.

![Figure 3](image)

As show in Figure 3b, the adhesive strength increased with the increase of duration of laser treatment and then reached a stable value. The adhesive strength markedly improved with the rise of laser treatment durations from 2 to 6 s, and then showed limited improvement with further rise of durations above 6 s. The durations of laser treatment brought in different amounts of energy transmitted to the pre-treated surfaces, resulting in different levels of pre-treatment qualities. We could obtain the theoretical energy density according to equation 1,

$$\text{Energy density} = \frac{P \cdot t}{S}$$  \hspace{1cm} (1)

where \(P\) represented the power of laser generator, \(t\) stood for the duration of laser treatment and \(S\) was the area of laser scanning. The energy densities with laser treatment durations were calculated from equation 1 as shown in Table 2.

| Duration/s | Scanning area/mm² | Energy density/J·mm⁻² |
|------------|-------------------|------------------------|
| 2          | 1050              | 0.0952                 |
| 4          |                   | 0.1905                 |
| 6          |                   | 0.2857                 |
| 9          |                   | 0.4286                 |
| 12         |                   | 0.5714                 |

In a summary, the use of pre-treatments did work on the removal of chemical conversion layer and improvement of adhesive bonding performance. And with laser treatment for 9 s, the adhesive strength was nearly equal to that with sand blasting. Due to the limited applicability of sand blasting used in pre-treatment of gluing, the laser treatment showed superiorities in convenience, cleanliness, precise processing area and little risk in work-pieces damage, which owned potential in popularization and application.
3.2. The morphologies of aluminum samples with pre-treatments

In order to investigate the adhesive strength improvement with pre-treatments, the morphologies of the samples were characterized by SEM as shown in Figure 4, for the certain morphology could provide superior interface to bonding. The blank sample showed a relatively flat and smooth surface. After

Figure 4. The morphologies of aluminum samples with pre-treatments: (a) blank, (b) chemical conversion, (c) grinding with 80 mesh sand paper, (d) sand blasting, laser treatment for (e) 2 s, (f) 4 s, (g) 6 s and (h) 9 s.
chemical conversion treatment, a uniform film with spreading cracks of less than 2 μm in width was formed, which was ascribed to chemical conversion film. The samples pre-treated with grinding and sand blasting showed intensely coarsening traces with staggered scratches and dense impact trails, respectively. The samples with laser treatments displayed the micromorphology with dispersed craters, where the craters with diameters of about 50 μm should result from the ablation of laser treatment.

Around the craters as shown in Figure 4e, the film with cracks appeared the same morphology to that in Figure 4b, which was attributed to the residual of uncleaned chemical conversion film. Although the chemical conversion film was barely found in macro view (Figure 2), the residual chemical conversion film could be observed by SEM in micro view. With the increase of duration of laser treatment, the area covered with craters increased and that with residual oxidation films decreased, which was clearly obtained from the surface morphologies and indicating the improvement of pre-treatment extents. The areas covered with craters apparently improved with the rise of laser treatment duration from 2 to 6 s, and then showed limited improvement with further rise of durations above 6 s, which corresponded well to the dependence of adhesive strength on durations as shown in Figure 3b. This phenomenon revealed that the longer duration of laser treatment with more amount of energy transmitted to the surface resulted in stronger pre-treatment, which made expected improvement on adhesive bonding performance.

Table 3. The surface roughness of aluminum samples with pre-treatments.

| No. | Pre-treatment                  | Ra/μm | Rz/μm |
|-----|--------------------------------|-------|-------|
| 1   | Blank                          | 1.116 | 7.836 |
| 2   | Grinding with 120 mesh sandpaper | 2.012 | 17.547|
| 3   | Grinding with 80 mesh sandpaper | 2.388 | 20.477|
| 4   | Sand blasting                  | 6.742 | 70.232|
| 5   | Laser treatment for 2 s        | 1.536 | 21.130|
| 6   | Laser treatment for 4 s        | 2.174 | 24.986|
| 7   | Laser treatment for 6 s        | 3.057 | 27.884|
| 8   | Laser treatment for 9 s        | 3.206 | 28.236|
| 9   | Laser treatment for 12 s       | 3.288 | 29.327|

Since the SEM got limited information about roughness, we adopted LCSM not only to observe the surface profiles, but also to obtain the average roughness (Ra) and maximum roughness (Rz). As defined in international standard of ISO 4287: 1997, Ra represented the extent of surface altitude undulation along X and Y directions and Rz equaled to the difference between maximum altitude and minimum altitude in roughness measuring area [13]. The surface roughness of aluminum samples with pre-treatments as listed in Table 3, and several typical surface profiles with pre-treatments were presented in Figure 5. The blank sample with a relatively low roughness undoubtedly appeared limited bonding strength, and the samples with pre-treatments showed obvious improvement in roughness which provided higher interface areas to bond, so that the adhesive strengths were significantly increased than that of blank sample. The samples pre-treated with grinding, sand blasting and laser treatment exhibited the morphologies of staggered scratches, dense impact trails and dispersed ablation craters, respectively, as shown in Figure 5. As applied with mechanical coarsening methods, the samples pre-treated with sand blasting showed much better bonding performance than that with grinding, because of a coarser surface developed during sand blasting which provide higher interface area to bond. The effect of different sandpaper mesh on adhesive strength could be explained in a similar way [1, 2]. With the increase in duration of laser treatment, the surface roughness increased, which coincided to the tendency of adhesive strength in Figure 3b. Although the pre-treatment of sand blasting developed much higher roughness than laser treatment, the sample with laser treatment for 9 s showed nearly equal adhesive strength to that with sand blasting. Since the sand blasting was a mechanical coarsening method which could barely improve the surface energy and wettability, the laser treatment ablating and even peeling off the outmost layer trough the mechanism including thermal vibration, vaporization, fusion and plasma stripping, which could not only roughen the surface.
but also increase the surface energy [5-7], as a result, the laser treatment made equal contribution to adhesive strength improvement to sand blasting though the roughening was much lesser.

Figure 5. Typical surface profiles of aluminum samples with pre-treatments: (a) grinding with 80 mesh sand paper, (b) sand blasting, laser treatment for (c) 2 s and (d) 9 s.

3.3. The wettability of aluminum samples with pre-treatments

Based on the analysis of surface morphology and roughness, we carried out contact angle measuring to characterize the surface wettability, in order to further understand the effect of laser treatments on adhesive bonding performance. The contact angles of water to aluminum samples with pre-treatments as shown in Figure 6. The blank sample showed a non-hydrophilicity with the contact angle of 92.3°. Compared with blank sample, the contact angle increased with pre-treated by grinding and sand blasting, for which the increase in surface roughness with mechanical coarsening was responsible referring to Wenzel’s studies [14]. The wettability turned to more hydrophobic after grinding and sand blasting, which meant few benefits on surface energy by mechanical coarsening method. However, for the samples pre-treated with laser scanning, the wettability to water had changed a lot compared with blank sample, which indicated great improvement in surface energy. The sample pre-treated with laser for only 2 s, the surface showed an excellent hydrophilicity with the contact angle to water of 14.4°. With the increase in duration of laser treatment, the hydrophilicity further enhanced, and the water drops was hardly observed in the video camera of optical contact angle meter with the duration rise to 9 s and above, which stood for the contact angles of nearly zero. These results had explained that laser treatment significantly improved the surface energy and enhanced the wettability [6, 10], by which made equal contribution to adhesive bonding strength to sand blasting though the roughening was much lesser.

With the results and discussion of surface morphology, roughness and wettability, we had found that the pre-treatment of laser scanning showed improvement not only on surface roughening but also on wettability, which contributed to the adhesive strength equaled to that of sand blasting.
4. Conclusions
We studied tensile shear strengths of epoxy adhesive bonding to 2A12 aluminum with preferred pre-treatments and investigated the influences of duration of laser treatment on the adhesive bonding strength to obtain the optimized extents of laser treatment. With the analysis of SEM, LCSM and contact angle measurements adopted to characterize the surface morphology, roughness and wettability, respectively, we discussed the effects of pre-treatments on the surface status and adhesive bonding strength. Then, we draw conclusions as following:

1) The adhesive strength significantly increased with surface roughening, and the pre-treatments of sand blasting and laser treatment resulted in much higher improvement on adhesive strength than that of grinding.

2) The durations of laser treatment brought in different amounts of energy transmitted to the pre-treated surfaces, resulting in different levels of pre-treatment qualities. Through the study on adhesive strength with laser treatment durations, we obtained an optimized laser treatment duration of 6 s with energy density of 0.2857 J·mm⁻², which could contribute to preferred adhesive bonding performance in our experiment conditions.

3) With the results and discussion of surface morphology, roughness and wettability, we had found that the laser treatment showed considerable improvement not only on surface roughening but also on wettability, which contributed to the adhesive strength equaled to that of sand blasting.

4) The laser treatment as pre-treatment before gluing showed superiorities in convenience, cleanliness, precise processing area and excellent improvement in adhesive bonding, which owned well potential in popularization and application.

References
[1] Borsellino C, Bella G Di and Ruisi V F 2009 Int. J. Adhes. Adhes. 29 36
[2] Yasmina B, Sami N, Salah M and Moez B S A 2016 Int. J. Adhes. Adhes. 67 38
[3] Shuochen C, Guangyao L and Junjia C 2018 Auto. Eng. 40 865
[4] Saleema E, Sarkar D K, Paynter R W, Gallant D and Eskandarian M 2012 Appl. Surf. Technol. 261 742
[5] Loutas T H, Kliafa P M, Sotiriadis G and Kostopoulos V 2019 Surf. & Coat. Technol. 375 370
[6] Yuyao L, Shun M, Qianming G, Yilun H, Jianming G, Ming Z, Bin L, Lei L, Guisheng Z and Daming Zhuang 2019 ACS Appl. Mater. Interfaces 11 22005
[7] Hongmin H, Jianping H, Kewei S and Lijun Z 2017 _J. Solid Rock. Technol._ **40** 779
[8] Wenfeng Y, Junlei L, Yu C, Yue L and Qingru T 2015 _Bull. Chin. Ceramic. Soc._ **34** 298
[9] Chang L, Wenfeng Y, Yi X, Shulun Z and Shaolong L 2019 _Aviat. Maint. & Eng._ **3** 32
[10] Jiangyou L, Minlin Z, Peixun F, Dingwei G and Hongjun Z 2015 _J. Laser Appl._ **27**, S29107
[11] Marco A, Gilles L, Franco F and Glaucio H P 2012 _Int J Adhes Adhes_ **9** 33
[12] Xiping L, Donghou X, Ningning G, , Zhenyu X, Linlin W and Weiping D 2019 _Mater. & Desig._ **179** 107875
[13] ISO 4287: 1997 Geometrical product specifications (GPS)-Surface texture: Profile method-
Terms, definitions and surface texture parameters.
[14] Wenzel R N 1949 _J. Phys. Chem._ **53** 1466