Effect of surface roughening of aluminum plates on the strength of bonds formed between aluminum and polyphenylene sulfide by thermosonic bonding

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Abstract. Thermosonic bonding of aluminum on polyphenylene sulfide was carried out in order to examine the effect of surface roughening of aluminum on the joint strength. Repeated chemical treatment of aluminum by immersion in aqueous sodium hydroxide and hydrochloric acid solutions increased its surface roughness ($R_a \approx 25 \mu m$) and surface area (~445% increase). Consequently, the bonding strength (~1.8N in average) was enhanced through anchoring effects.

1. Introduction
Polyphenylene sulfide (PPS) is one of the most promising heat- and chemically resistant super-engineering thermoplastics. Adhesive bonding is generally adopted for the joining this polymer to surface-treated metals for use in automotive and electronics industries [1]. The use of adhesives, however, has certain problems, such as low working efficiency, detrimental effect on environment and workers, and unreliability of joint over a long term due to chemical degradation. Considering these issues, we conducted thermosonic bonding, whose primary joining principle is the anchoring effect obtained by plastic resin deformation and its penetration into metal surface pits. For this reason, chemical etching in alkaline/acid solutions is one of the simpler choices of metal surface roughening. For example, upon immersion in an active base of pH 13, such as an aqueous solution of sodium hydroxide [2], aluminum, even in the presence of its stable passivating native oxide layer, is etched uniformly to a matte finish, thereby introducing a high density of nucleation sites for pits on the surface [3]. When subsequently immersed in a solution of hydrochloric acid, aluminum is etched, as described by the following reaction equations.

$$\text{Al}_2\text{O}_3 (s) + 2\text{NaOH}(aq.) + 3\text{H}_2\text{O} \rightarrow 2\text{Na}[\text{Al(OH)}_4] (aq.) \quad (1)$$

$$2\text{Al} (s) + 2\text{NaOH}(aq.) + 6\text{H}_2\text{O} \rightarrow 2\text{Na}[\text{Al(OH)}_4] (aq.) + 3\text{H}_2 (g) \quad (2)$$

Since the product of both reactions (1) and (2), sodium aluminate, is soluble in the basic solution of sodium hydroxide [2], aluminum, even in the presence of its stable passivating native oxide layer, is etched uniformly to a matte finish, thereby introducing a high density of nucleation sites for pits on the surface [3]. When subsequently immersed in a solution of hydrochloric acid, aluminum is etched, as described by the following equation.

$$2\text{Al} (s) + 6\text{HCl} (aq.) \rightarrow 2\text{AlCl}_3 (aq.) + 3\text{H}_2 (g) \quad (3)$$
This reaction occurs rather preferentially at the pitting nucleation sites related to surface flaw distribution [4], and proceeds anisotropically, resulting in the formation of rectangular etch pits. When again subjected to the NaOH solution, the step described by eq. (2) occurs again, and the rectangular etch pits grow rather rapidly to take on round-shaped crater forms through rapid anodic dissolution due to the exposure of the cathodic second phase (iron-rich precipitates in case of A1050) on the aluminum surface [5]. By repeating the alternating alkaline/acid immersions, anomalous roughening, characterized by a very large increase in the size of the aluminum surface craters, can be produced. In this study the effect of surface roughening of aluminum plates on the strength of the bonds formed between aluminum and PPS by thermosonic bonding was investigated.

2. Experimental procedure
Surface roughening of aluminum was carried out as pretreatment under the following three conditions: (treatment C1) immersion in sodium hydroxide (5 wt% aq. solution) for 120 s at 40 °C and then in hydrochloric acid (3 wt% aq. solution) for 300 s at 40°C; (treatment C2) dipping in sodium hydroxide (5 wt% aq. solution) for 150 s at 40 °C after C1; and (treatment C3) dipping in sodium hydroxide (5 wt% aq. solution) for 300 s at 40 °C after C1. After each treatment, a pair comprising an aluminum plate (A1050P H24, 5 mm square and 0.5 mm thick) and a PPS plate (1 mm in thickness) was used for the joining process. Physical properties of PPS are shown in Table 1. After subjecting to the surface treatments, the aluminum plates were bonded on to PPS plates using an ultrasonic bonder by solid-state bonding. The bonding tool head vibrates horizontally at a frequency of 50 kHz, and is capable of performing thermo-compression as shown in Figure 1. The bonding conditions were fixed as follows: temperature of 200 °C, pressure of 12 MPa, bonding time of 2.5 s, and an ultrasonic vibration amplitude of 6 μm. The tool vibration amplitude was controlled through closed-loop feedback during the bonding cycle. Shear tensile testing of the joined specimen was conducted for the evaluation of the bond strength at a crosshead speed of 0.1 mm/s. The tensile direction was set parallel to the direction of ultrasonic vibration.

### Table 1. Physical properties of polyphenylene sulfide.

| Property                      | Value               |
|-------------------------------|---------------------|
| Melting point (°C)            | 250                 |
| Glass transition temperature (°C) | 87                  |
| Specific gravity              | 1.50                |
| Rockwell hardness            | R118                |
| Tensile strength (MPa)       | 130                 |

![Figure 1. Schematic illustration of the thermosonic bonding tool, which vibrates horizontally and is capable of performing thermo-compression.](image)
Quantitative topological characterization of the aluminum surface was conducted by scanning laser microscopy (Keyence, VK-5800). Surface roughness $R_a$ ($\mu m$) and surface area $S$ ($\mu m^2$), as observed in the images, were calculated using the system software provided with the microscope from the measured topographical array data. The percentage increase in surface area, $\alpha$, was evaluated using the following equation.

$$\alpha (%) = \left( \frac{S_1 - S_0}{S_0} \right) \times 100$$

where $S_1$ and $S_0$ are the surface areas of aluminum after and before chemical etching, respectively.

3. Results and discussion

Aluminum sample surfaces after the chemical etching treatments are shown in Figure 2. Surface evaluation results of etched aluminum are presented in Table 2. For C1 treatment, small pits with a low density (pit ratio 13.8%) were formed. The pits were either small rectangles or rectangular aggregates with sharp edges. However, the majority of almost surface area remained unaffected, showing a flat topology with rolling scratches similar to surface of as-received aluminum. On the other hand, hemispherical pits with a high density (pit ratio > 80%) were formed after both C2 and C3 treatments. The average pit diameter of pits was several tens of micrometers, comparable to the surface roughness $R_a$. Moreover, the percentage increase in surface area, $\alpha$, increased linearly with $R_a$ up to a maximum of 445% ($\alpha \approx 20.3 \times R_a - 54.7$). It is thus concluded that the aluminum surface roughness ($R_a \approx 25 \mu m$) can be increased with keeping almost linear relation to the increase in surface area by simple repeated alkaline/acid chemical etching.

![Figure 2](image-url)

*Figure 2.* Aluminum surface etched by the following chemical treatments: C1 ((a), (d)); C2 ((b), (e)); and C3 ((c), (f)).
Table 2. Characterization results of aluminum surface after chemical treatments.

| Treatment | Surface roughness $R_a$ (μm) | Percentage increase in surface area, $\alpha$ | The average diameter of the pit (μm) | Pit ratio in the observed range (%) |
|-----------|-------------------------------|-----------------------------------------------|-------------------------------------|-----------------------------------|
| As received | 3.2                           | 0                                             | -                                  | -                                 |
| C1        | 4.8                           | 38                                            | -                                  | 13.8                              |
| C2        | 15.5                          | 292                                           | 12-14                              | 80.9                              |
| C3        | 25.5                          | 445                                           | 20-25                              | 93.5                              |

Figure 3. Effect of chemical treatments on the surface roughness $R_a$, surface increase ratio $\alpha$, and shear strength.

The effect of the chemical treatments on the shear strength of the bonded joints is shown in Figure 3. In the figure, $R_a$ (light gray) and $\alpha$ (dark gray) are also represented as solid bars. The joints made using specimens of as-received aluminum and those subjected to C1 treatment, both of which have small values of $R_a$ and $\alpha$, showed a small average shear strength value of less than 0.5 N. Because of the small density of the etch pits, as shown in Figure 2(a), it is thought that sufficient anchoring onto aluminum pits could not be achieved. It should also be pointed out that using too large an ultrasonic amplitude (6 μm) compared to the surface roughness (3.2 μm) was ineffective for proper bonding. In such cases of a large difference between amplitude and roughness, it is very probable that the resin flows in and out of the small aluminum pits repeatedly, and consequently, no stable anchor bonds are formed, this effect produced being much like the grinding of PPS. In contrast to the C1 treatment, the C2 and C3 treatments caused an increase of the average shear strength, approximately 1.2 N and 1.8 N, respectively. It is assumed that the increase of the average shear strength is attributable to the surface roughening of aluminum to a surface roughness value far greater than the amplitude of the ultrasonic vibration. Nevertheless, the minimum values of shear strength obtained through C2 and C3 treatments merely reached approximately 0.5 N, which is almost similar to the average value obtained through C1 treatment. Since no interfacial interatomic chemical reaction or inter diffusion can be expected from such low-temperature thermosonic bonding [6], the variations in bonding strength is solely attributable to the variations in the ‘interfacial topology’ between aluminum and PPS.

In order to elucidate the origin of such bonding characteristics, the bonded aluminum and PPS plates of a few bonded samples were separated, and the previously bonded areas were studied. As shown in Figures 4(a) and (b) for the case of C3 treatment, it was confirmed that very similar surface topology was observed on both aluminum and PPS surfaces at the micrometer scale. It clearly shows that the
localized deformation of PPS at the aluminum pits was achieved through ultrasonic vibration amplitudes smaller than pit sizes even under low bonding pressures (12 MPa) below the melting point of PPS. The horizontal vibration of the pit peripheral ridges might have caused localized plastic deformation of the resin. In such a case, the thermosonic energy (the sum of ultrasonic energy and thermal energy) could have been effectively absorbed only by the PPS surface, leading to bonding. However, it should be mentioned that there existed a certain amount of topology mismatch between PPS and aluminum. The PPS image was not sharp, and its surface was not accurately identical to that of aluminum, especially around the smaller pits across the rugged area (light gray zone). This observation leads to the inference that the existence of non-bonded gaps at the interface is the potential reason for the variations in the bonding strength.

The cross-sectional optical microphotograph of a bonded specimen subjected to C3 treatment is shown in Figure 5. The resin remains adhered to the aluminum surface despite the rough interface. The increase in aluminum surface roughness and the resultant increase in the interfacial area between aluminum and PPS together caused the average bonding strength to increase. However, non-bonded small voids at the interface were also observed, such as the one shown by white arrow in the figure. This incomplete intrusion of resin might be another cause of the variations in the shear strength in the case of C2 and C3 treatments, since the existence of voids at the interface increases the local stress, and initiates cracks during shear testing. It is suggested that the wider variation of the shear bonding strength was caused by the uncontrolled variation in the non-bonded interface defects. Further study on the effects of surface pretreatment through more controlled roughening by an electrochemical approach as well as on the effects of the thermosonic process parameters shall be undertaken for improving bonding quality.

![Figure 4](image1.png)

**Figure 4.** Bonded C3-treated pair of (a) aluminum surface and (b) PPS (mirror images).

![Figure 5](image2.png)

**Figure 5.** Cross-sectional optical microphotograph of the bonded interface of aluminum (C3 treatment) and PPS. A pit (white arrow) remains as an interfacial void.
4. Conclusions
Thermosonic bonding of aluminum on PPS resin was examined through the use of a horizontally vibrating tool head. The aluminum surface treatment by the repeated alternated immersion in NaOH and HCl aqueous solutions increased the surface roughness ($R_a \sim 25\mu m$) with a linear increase in surface area ($\alpha \sim 445\%$) through the formation of a high density of micro pits. The average bonding strength increased up to 1.8 N with increasing surface roughness. The application of ultrasonic vibration was effective due to the thin plastic, but not severe, deformation and anchoring of PPS. The non-bonded zone caused by low roughness (< ultrasonic vibration amplitude) and incomplete anchoring are supposed to be the causes of the wide scatter of the bonding strength.

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