International Journal of ELECTROCHEMICAL SCIENCE www.electrochemsci.org

Short Communication

# Enhanced Diffusion Bonding Between High Purity Aluminum and 6061 Aluminum by Electrolytic Polishing Assistance

May-Show Chen<sup>1,2</sup>, Fang-Yu Fan<sup>3</sup>, Chung-Kwei Lin<sup>3,4,\*</sup>, Chien-Chon Chen<sup>4,5,\*</sup>

<sup>1</sup> School of Oral Hygiene, College of Oral Medicine, Taipei Medical University, Taipei 11031, Taiwan

<sup>2</sup> Department of Dentistry, Taipei Medical University Hospital, Taipei 11031, Taiwan

<sup>3</sup> School of Dental Technology, College of Oral Medicine, Taipei Medical University Taipei 11031, Taiwan

<sup>4</sup> Research Center for Biomedical Devices and Prototyping Production, Taipei Medical University, Taipei 11031, Taiwan

<sup>5</sup> Department of Energy Engineering, National United University, Miaoli 36003, Taiwan

\*E-mail: <u>chungkwei@tmu.edu.tw</u>, <u>chentexas@gmail.com</u>

Received: 21 March 2016 / Accepted: 15 April 2016 / Published: 4 May 2016

The rolling trace on aluminum alloys usually affects the post surface treatments of high quality aluminum alloy products, such as medical supplies, 3C products, sport equipment, automobile parts, and aircraft parts. In order for aluminum alloys to have good mechanical properties and a high quality of surface smoothing and brightness, an aluminum alloy (6061 Al) substrate surface is bonded with high purity aluminum (5N Al). This work reports the development of a cost-effective way to join the interface between 5N Al and 6061 Al by electrolytic polishing and thermal pressure diffusion methods. Electrolytic polishing can smooth the surfaces of both 5N Al and 6061 Al, dissolve partial precipitates, form small pits, and generate an Al-rich zone on the 6061 Al surface. The small pits, smooth surfaces, and Al-rich zone enhance surface bonding in the thermal pressure diffusion process.

Keywords: Aluminum alloy, Pure aluminum, Joining , Diffusion, Interface

## **1. INTRODUCTION**

6061 aluminum alloy (6061 Al) is a precipitation-hardened aluminum alloy that has mechanical properties superior to those of pure aluminum. It is one of the most common alloys of aluminum used in 3C (computer, communication, and consumer electronics) products. High quality surface treatments of 6061 Al include electrolytic polishing or anodization to enhance the value and lifetime of 3C

products. However, it is difficult to obtain a reflective surface on 6061Al by electrolytic polishing because its major alloying elements are magnesium (Mg) and silicon (Si), which have different reduction potentials. The basic mechanical proporties of 6061 Al included density (2.70 g/cm<sup>3</sup>), tensile strength: yield (270 MPa ) tensile strength: (310 MPa), elongation at break (15 %), thermal conductivity (170 W/m-K), fatigue strength (97 MPa), and specific heat capacity (900 J/kg-K) [1].

Pure Al is a soft, nonmagnetic, ductile metal that is highly reflective, especially after surface electrolytic polishing, and unlike 6061 Al, it has no rolling lines. High quality 3C products usually require a substrate with good mechanical properties and a smooth, reflective surface and surface cover of a transparent corrosion- and wear-resistant film.

Aluminum is also an active metal that naturally forms an oxide layer on the surface in ambient surroundings or aqueous solution. This natural tendency for oxidation interferes with efforts to add a coating of Al film on the Al substrate by normal methods. In the literature, several methods of deposition or coating Al on the specific substrate have been reported, examples being electrodeposition [2, 3, 4], vapor deposition [5], and cold spray [6, 7]. However, these approaches are expensive because of the expensive equipment, requirement of an ionic liquid, and thinness of the film thus formed.

Electrolytic polishing (EP) was invented by Pierre Jacquet of France in 1929 and was first successfully applied to metallographic sample preparation in 1935 [8]. It is best suited for pure metals and single-phase alloys. On the contrary, multiphase alloys are often quite difficult because of differences in the polishing rates of the constituents. There are some important conditions for the EP. Examples of those conditions are initial surface roughness, electrolytic composition, electrolytic temperature, and stirring rate of electrolyte, applied voltage, current density, and polishing time. If the roughness of initial surface is too coarse, EP time will be long, with excessive metal removal and waviness. For the voltage, the applied voltage is determined by the curves of voltage vs. current density (I-V curve).

It would be beneficial to combine the good mechanical properties of 6061 Al and good surface treatment of pure Al. In order to reduce the cost of the Al-6061 Al joining process, we have developed a process for bonding a thicker pure Al foil or sheet with 6061 Al by surface electrolytic polishing and thermal pressure diffusion methods. Bonding a thicker pure Al sheet to the 6061 Al surface can allow further surface treatments such as electrolytic polishing to produce a highly reflective surface, anodization to produce a hard coating, and adding a structured oxide film by anodic aluminum oxide (AAO) method.

#### 2. EXPERIMENTAL PROCEDURE

The thermal diffusion bonding materials were high purity Al (99.999%, 5N Al) disks of 2 inches in diameter and 0.8 mm in thickness, and Al alloy (Al-Mg-Si; 6061 Al) disks of 2 inches in diameter and 100 mm in thickness. In order to understand the surface roughness after surface treatments, the surfaces of the 6061 Al samples were treated with various methods: waterproof silicon carbide polishing with paper up to #2000, sodium hydroxide soaking (10 wt.% NaOH), chemical

polishing (60 vol.%  $H_2SO_4 + 30$  vol.%  $H_3PO_4 + 10$  vol.%  $HNO_3$  at  $120^{\circ}C$  for 1 min), and electrolytic polishing.

In order to obtain clean and smooth surfaces, the 5N Al and 6061 Al surfaces were ground with sandpaper and electrolytically polished. The surfaces were then bonded tightly using a thermal diffusion method. Finally, a hard oxide film or an AAO film with an ordered channel structure was formed on the 6061 Al surface. The experimental conditions of electrolytic polishing, thermal diffusion bonding, and the AAO process are described as follows:

For the electrolyte temperature and stirring rate, when the bath temperature or stirring rate rises, the resistance of the bath decreases and the potential required to produce the plateau current density decreases. In addition, the bath viscosity decreases, making it more difficult to maintain a viscous anode layer.

Aluminum surface pretreatments: The Al surface was first mechanically ground using a series of sandpaper grades up to #2000. Then the Al surface was further polished by electrolytic polishing (15 vol.% HClO<sub>4</sub> + 15 vol.% (CH<sub>3</sub>(CH<sub>2</sub>)<sub>3</sub>OCH<sub>2</sub>CH<sub>2</sub>OH + 70 vol.% C<sub>2</sub>H<sub>6</sub>O, at 42 V for 5 min).

Bonding of high purity Al to 6061 Al: The electro-polished surfaces of high purity Al and 6061 Al were placed in contact and set in a diffusion mold under thermal diffusion conditions of 450 kgf/cm<sup>2</sup> pressure at 320 °C for 1 h.

When anodized in an acidic electrolyte and controlled in the suitable conditions, aluminum forms a porous oxide with very uniform and parallel cell pores. Each cell contains an elongated cylindrical sub-micron or nanopore normal to the aluminum surface, extending from the surface of the oxide to the oxide/metal interface, where it is sealed by a thin barrier oxide layer with approximately hemispherical geometry. Its structure is described as a close-packed array of columnar cells. The pore diameters and pore densities of anodic alumina film can be finely controlled by appropriately carrying out the electrochemical process parameters [9, 10].

Ordered pore structure of anodic aluminum oxide (AAO): AAO film was made by anodization. The steps included: (a)  $1^{st}$  anodization (1.5 vol.% H<sub>3</sub>PO<sub>4</sub>, at 200 V, 3 wt.% C<sub>2</sub>H<sub>2</sub>O<sub>4</sub> at 40V, or 10 vol.% H<sub>2</sub>SO<sub>4</sub> at 18V), (b) removal of  $1^{st}$  anodization film (1.8 wt.% CrO<sub>3</sub> + 6 vol.% H<sub>3</sub>PO<sub>4</sub> + 92 vol.% H<sub>2</sub>O at 70 °C), (c)  $2^{nd}$  anodization 1.5 vol.% H<sub>3</sub>PO<sub>4</sub>, at 200 V, 3 wt.% C<sub>2</sub>H<sub>2</sub>O<sub>4</sub> at 40V, or 10 vol.% H<sub>2</sub>SO<sub>4</sub> at 18V), and (d) pore widening (5 vol.% H<sub>3</sub>PO<sub>4</sub>, for 10 min, 1.5h, or 4h). The details of the ordered structure of the AAO process can be found in our previous articles [11, 12, 13].

The experimental chemicals (sodium hydroxide (NaOH), perchloric acid (HClO<sub>4</sub>), ethylene glycol butyl ether (CH<sub>3</sub>(CH<sub>2</sub>)<sub>3</sub>OCH<sub>2</sub>CH<sub>2</sub>OH), ethanol (C<sub>2</sub>H<sub>6</sub>O), chromium trioxide (CrO<sub>3</sub>), phosphoric acid (H<sub>3</sub>PO<sub>4</sub>), sulfuric acid (H<sub>2</sub>SO<sub>4</sub>), and oxalic acid (C<sub>2</sub>H<sub>2</sub>O<sub>4</sub>)) were purchased from Merck in reagent grade. The microstructure and composition of the fabricated samples were studied using optical microscopy (Nikon LV 150) and scanning electron microscopy (SEM, JEOL 7400 and 6510).

#### **3. RESULTS AND DISCUSSION**

Aluminum alloy joints with metals has been widely studied; for example, 6061 aluminum alloy joints with Al–10.8Si–10Cu and Al–9.6Si–20Cu filler metals at 560 °C [14], hard carbon particulate

reinforced 6061 Al alloy by microwave sintering method [15], and by friction stir welding (FSW) [16]. According to the literature [17], the secondary phases identified thus far in 6061 Al include Al<sub>3</sub>Fe,  $\alpha$ -(Fe,Cr,Mn)3SiAl<sub>12</sub>,  $\beta$ -(Fe,Cr,Mn)2Si<sub>2</sub>Al<sub>9</sub>, Si, Mg<sub>2</sub>Si and  $\pi$ -(Fe,Cr,Mn)Mg<sub>3</sub>Si<sub>6</sub>Al<sub>8</sub>.



**Figure 1.** Optical microstructures of 6061 Al after (a) grinding with #2000 sandpaper, (b) soaking in alkaline solution of 10 wt.% NaOH, (c) chemical polishing in 30 vol.% H<sub>3</sub>PO<sub>4</sub> solution, (d) electrolytic polishing in 15 vol.% HClO<sub>4</sub> solution.

The transition and precipitated equilibrium phases are  $\alpha_{sss} \rightarrow GP_{zones} \rightarrow \beta' \rightarrow \beta'' \rightarrow \beta$  phase [18]. These phases are present as precipitates in the aluminum matrix. Figure 1 shows optical microstructure images of 6061 Al after different surface treatments: (a) The scratches and pits remained after grinding with #2000 SiC paper, (b) surface roughness increased and obvious scratches and pits appeared after soaking in 10 wt.% NaOH solution, (c) precipitates dissolved, big pits formed, and grain boundaries presented after chemical polishing in 30 vol.% H<sub>3</sub>PO<sub>4</sub> solution, (d) the surface was smooth, the precipitates were replaced with small pits, and grain boundaries presented after electrolytic polishing in 15 vol.% HClO<sub>4</sub> solution.

In order to bond 5N Al to 6061 Al substrate, we designed a thermal pressure diffusion mold. The 5N Al foil and 6061 Al sheet were set on the thermal diffusion mold, and constant pressure was applied by screws and hydraulic machine. Figure 2 shows the thermal diffusion mold and samples. (a) Schematic diagram of thermal diffusion mold; the structure includes the top-mold and bottom-mold as the mold body, screws and nuts for maintaining constant pressure, and samples of 5N Al /6061 Al set

between top-sheet and bottom-sheet, (b) actual image of thermal diffusion mold made of 304 stainless steel, (c) 2-inch diameter sample of 0.8 mm thick 5N Al bonded on 100 mm thick 6061 Al substrate by thermal diffusion method, (d) schematic diagram of 6061 Al substrate with 5N Al foil and ordered channel formation after thermal diffusion method and anodization process.



**Figure 2.** Schematic diagrams and images of 5N Al diffused on the 6061 Al substrate; (a) schematic diagram of 5N Al foil and 6061 Al sheet set on the thermal diffusion mold, (b) actual image of thermal diffusion mold, (c) digital camera images of 5N Al foil diffused on the 6061 Al sheet, (d) schematic diagram of 6061 Al substrate with 5N Al foil and ordered channel formation.



**Figure 3.** Optical micro images of (a) microstructure of 5N Al, (b) microstructure of 6061 Al, (c) 6061 Al before mechanical polishing, after mechanical polising, and after electrolytic polishing, and 5N Al after electrolytic polishing, (d) image of electrolytically polished 5N Al diffused on 6061 Al.

Figure 3 shows optical microstructure images of 5N Al, 6061 Al, and 5N Al/6061Al after electrolytic polishing. (a) 5N Al presents an optical-grade flat surface with larger grains, (b) 6061 Al presents precipitates, small grains, and pits, (c) digital camera images of 6061 Al showing a rough surface before mechanical polishing; after mechanical polishing, a smooth surface; after electrolytic polishing, a smooth and reflective surface; and after electrolytic polishing, a good quality surface of 5N Al, (d) image of electrolytically polished 5N Al diffused on 6061 Al, showing an obviously good quality surface of 5N Al compared to the rolling traces of the 6061 Al surface.



**Figure 4.** SEM images of AAO microstructures formed on the pure Al of 6061 Al substrate. Pore sizes of (a) 15 nm, (b) 30 nm, and (c) 450 nm, and (d) straight tube structure.

Numerous researchers have focused on the high purity (99.999%) of an aluminum substrate, the control of applied voltage, electrolyte pH, and the composition of the solution, pore widening and compensation processes to fabricate a regular array of nanopores on an alumina film [19, 20, 21, 22] by two-step anodization process [23, 24]. Because 6061 Al contains Si and Mg elements that easy to

form Al<sub>3</sub>Fe,  $\alpha$ -(Fe,Cr,Mn)3SiAl<sub>12</sub>,  $\beta$ -(Fe,Cr,Mn)2Si<sub>2</sub>Al<sub>9</sub>, Mg<sub>2</sub>Si and  $\pi$ -(Fe,Cr,Mn)Mg<sub>3</sub>Si<sub>6</sub>Al<sub>8</sub> in the Al matrix [18] the disordering pore structure can forming on the Al surface after anodization [25].

In order to form a high purity Al film on the good mechanical quality of Al alloy there are mrthods has been proposaled; for example electrodeposition of aluminum on aluminum surface from molten salt [26], pulse current electrodeposition of Al from ionic liquid [27, 28]. However, the high temperature of molten salt and expensive of ionic liquid may restrict the technologies using in the industry.

An anodic film with an ordered pore structure can form on the pure Al, but a disordered anodic film forms on Al alloys such as 6061 Al. Figure 4 shows the ordered pore structure of AAO films formed on the pure Al surface of 6061 Al substrate by diffusion bonding method: (a) pore size of 15 nm after anodization at 18V in 10 vol. %  $H_2SO_4$ ; (b) pore size of 30 nm and 120 nm after anodization at 60 V and then 40 V in 3 wt. %  $C_2H_2O_4$ ; (c) pore size of 450 nm after anodization at 200 V in 1.5 vol. %  $H_3PO_4$ ; (d) pore size of 450 nm AAO with straight tube structure. The ordered AAO enhances the sealing and drying efficiency and the hardness, enhancing the color saturation, corrosion resistance, and wear resistance of the surface. In further applications, the ordered pore structure can act as a template for storage of small materials such as chemical elements, metals, nanoparticles, or drugs.

### 4. CONSLUSIONS

The novelty in this paper included: (1) A cost-effective way to join the interface between 5N Al and 6061 Al, (2) Bonding pure Al with 6061 Al by electrolytic polishing and diffusion methods, (3) The smooth Al and 6061 Al surfaces allows easy interface bonding under diffusion, and (4) This technology can also be scaled up to industrial application.

In summary, 5N Al has been bonded with 6061 Al by electrolytic polishing and thermal pressure diffusion methods. The electrolytic polishing provides smooth 5N Al and 6061 Al surfaces, and the pitting of the 6061 Al surface allows easy interface bonding under thermal pressure diffusion. This workpiece is applicable to high temperature use or post treatment; for example, the workpiece can be treated with micro-arc oxidation because the clean interface of 5N Al/ 6061 Al has no solder. This technology can also be scaled up to industrial application because the advantages of this method include large-scale, easy operation processes; automation; mass production; and low equipment costs.

#### ACKNOWLEDGEMENTS

The authors would like to thank Taipei Medical University for financial support this work under grant no. 102TMU-TMUH-12.

#### References

1. Aluminum Association Completes 2013 Edition of Aluminum Standards and Data <u>http://www.aluminum.org/news/aluminum-association-completes-2013-edition-aluminum-standards-and-data#sthash.3T2gx4kt.dpuf</u>

- 2. T. Jianga, M.J.C. Brymb, G. Dubéb, A. Lasiaa, and G.M. Brisarda, *Surf. Coat. Technol.*, 201 (2006) 10.
- 3. T.C. Huynh, Q.P.D. Dao, T.N. Truong, N.G. Doan, and S.L. Ho, *Environ. and Pollut.*, 4 (2014) 59.
- 4. H.A.M. Abdel, and M Ichimura, *Mater Lett.*, 130 (2014) 26.
- M. Veith, C. Petersen, O.C. Aktas, W. Metzger, M. Oberringer, T. Pohlemann, M. Müller, and S. Gerbes, *Mater Lett.*, 62 (2008) 3842.
- 6. Q. Wang, N. Birbilis, and M.X. Zhang, Mater Lett., 65 (2011) 1576.
- 7. A. Moridi, S.M.H. Gangaraj, M. Guagliano, and M. Dao, Surf. Eng., 36 (2014) 369.
- 8. F. George and V. Voort, Metallography Principles and Practice, McGraw-Hill book company, 1984.
- 9. C.A. Huber, and T.E. Huber, Science, 263 (1994) 800.
- 10. D. Routkevitch, and J.M. Xu, Trans. Electron. Dev., 43 (1996) 1646.
- 11. J.S. Lin, S.H. Chen, K.J. Huang, C.W. Hun, and C.C. Chen, Atlas J. Mater. Sci., 2 (2015) 65.
- 12. C.Y. Chen, C.W. Hun, S.F. Chen, C.C. Chen, J.S. Lin, S.S. Johnson, N. Noel, N. Juliely, and Z.P. Luo, *Rev. Nanosci. & Nanotech.*, 4 (2015) 26.
- 13. C.C. Chen, D. Fang, and Z.P. Luo, Rev. Nanosci. & Nanotech., 1 (2012) 229.
- 14. S.Y. Changa, L.C. Tsaob, T.Y. Lia, and T.H. Chuang, J. Alloys Compd., 488 (2009) 174.
- 15. R.R. Zhenga, Y. Wua, S.L. Liaoa, W.Y. Wangb, W.B Wangb, and A.H. Wang, *J. Alloys Compd.*, 590 (2014)168.
- 16. N.Z. Khan, A.N. Siddiquee, Z.A. Khan, and S.K. Shihab, J. Alloys Compd., 648 (2015)360.
- 17. S. Rodríguez, E. Silva, R. Haddad, and L. Lanzani. SAM-CONAMET/SIMPOSIO MATERIA, 06-46 (2003) 585.
- 18. R.X. Li, R.D. Li, Y.H. Zhao, L.Z. He, C. Li, H.R. Guan, and Z.Q. Hu, *Mater. Lett.*, 58 (2004) 2096.
- 19. H. Masuda and H. Yamada, Appl. Phys. Lett., 71 (1997) 2770.
- 20. H. Masuda and F. Hasegwa, J. Electrochem. Soc., 144 (1997) 127.
- 21. H. Masuda and K. Fukuda, Science, 268 (1995) 1466.
- 22. H. Masuda and M. Yotsuya, Appl. Phys. Lett., 78 (2001) 826.
- 23. R. J. Tonucci and B, L. Justus, Science, 258 (1992) 783.
- 24. R. Charles and H. Martin, Chem. Mater., 8 (1996) 1739.
- 25. T.C. Cheng and C.C. Chou, J. Nanomaters., 2015 (2015) 371405.
- 26. W. Huang, X. Xia, B, Liu, Y, Liu, H. Wang, and M.A. Naiheng, Acta Metall. Sin., 24 (2011) 443.
- 27. B. Li, C. Fan, Y. Chen, J. Lou, L, Yan, Electrochim. Acta, 56 (2011) 5478.
- Z. Lipeng, W. Enqi, M. Jiechen, Y. Xianjin, W. Qiannan, Y. Lina, and Z. Zengdian, J. Nanoscience and Nanotechnology, 14 (2014) 6287.

© 2016 The Authors. Published by ESG (<u>www.electrochemsci.org</u>). This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution license (http://creativecommons.org/licenses/by/4.0/).