A mathematical modelling of preheated accumulative roll bonded Al-Al$_2$O$_3$ composite sheet

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Abstract

Accumulative roll bonding (ARB) technique is used in this paper to produce aluminum/alumina composite sheets. Alumina content was added as 1,3 and 5wt%. The produced Al/Al$_2$O$_3$ composite sheets are piled up and processed by accumulative roll bonding (50% reduction) after preheating at 280 °C with different regimes (2–8 cycles). Statistical design analysis was applied to examine the effects of alumina content and no. of accumulative roll bonding cycles on the ultimate tensile strength for aluminum/alumina composite sheets. Empirical formulas were deduced to recognize key parameters that controlling tensile behavior. XRD detection was carried out to explore dominant planes controlling plasticity Al/Al$_2$O$_3$ composites. In general, addition of alumina and proceeding different cycles increases strength. FE-SEM microstructure showed that alumina plays important roll on the aluminum sheets during ARB process where the metal of aluminum flow among them producing highly sheared matrix.

1. Introduction

Aluminum/alumina (Al/Al$_2$O$_3$) composite are considered new advanced materials in industries because of their light weight, good wear and corrosion resistance, high strength and high modulus of elasticity and low coefficient of thermal expansion. Several methods are used to fabricate Al/Al$_2$O$_3$ composite such as powder metallurgy [1], squeeze casting and spray forming [2, 3] accumulative roll bonding (ARB) [4] equal channel angular pressing (ECAP) [5] and multi-axial free forging. ARB is known as kind of severe plastic deformation (SPD) methods [6]. The ARB method is considered as easy production and based on cold rolling process [7]. ARB technique is one kind of the (SPD) mechanism in which large plastic strain is enacted into the material [8]. In order to attain an ultra-fine-grained metal with excellent mechanical properties. In Al/Al$_2$O$_3$ composite, fine particles of Alumina (<50 μm) is utilized to produce Al/Al$_2$O$_3$ composite. Fabricating of Al/Al$_2$O$_3$ composite using ARB process cold at room temperature exhibits Al/Al$_2$O$_3$ sheets of very weak bonding strength due to severe strain hardening of metal layers. For example, large number of cracks begin to nucleate and propagate during cold rolling passes [9]. Reduction in thickness and preheating temperatures are key parameters that control the preconditioning for successful bonding of Al/Al$_2$O$_3$ layers. At higher reduction of thickness, the bonding strength between two layers of composite rises[10]. Very few papers demonstrate the ARB at higher temperature. It was found that the peeling force (Adhesive Bonding Force) of composite layers enhances [11]. The target of this research is to manufacture Al/Al$_2$O$_3$ composite sheet containing different Alumina using preheating ARB Process (at 280 °C). Also, to correlate the relationship between Alumina content and number of cycles on the ultimate tensile strength (UTS) using Experimental Design Technique (Statistical design analysis).
2. Experimental work

The aluminum sheets dimensions were of length, width and thickness 330 mm × 150 mm × 1.5 mm strips, respectively. The direction of the cutting of strip was parallel to the sheet rolling direction. The quality of surface preparation is important in ARB. Surface preparation was carried out to increase the adhesion aluminum (AA1050) sheets. It can be done chemically and mechanically. Chemical cleaning (degreasing) entails the removal of dirt, oil, grease and other external materials with organic solvents as acetone solution. Al sheets were subjected to mechanical cleaning (wire brushing). Specification of scratch brush used in this study with a 90 mm diameter stainless-steel circumferential brush with 0.27 mm wire diameter, 35 mm wire length and peripheral speed of 2000 rpm. After surface preparation, Al₂O₃ particles were uniformly diffused between the two strips of aluminum sheets with different content of alumina (1,3 and 5wt%). Al/Al₂O₃ sheets were then stacked over each other and fastened at both ends by steel wires. The experiment pays minute attention to proper alignment of the two strip surfaces before rolling. Assembled Al/Al₂O₃ sheets containing different Al₂O₃ content (1,3 and 5wt%) were preheated at 300 °C for 5 min and then rolled for single pass with 67% reduction in thickness (without any lubrication) to 1mm strip thickness and air cooled. This produced sandwiches of Al/Al₂O₃ composite (containing different concentration of alumina) is called (Cycle 0). To remove work hardening from previous steps and to improve bonding strength of the cycle 0 sandwich, Al/Al₂O₃ composites sheets then (1, 3 and 5wt% Al₂O₃) were fully annealed at 400 °C for 60 min, and then cooled in the furnace. The fully annealed sandwiches were cut in perpendicular to the ARB direction. According to the number of assembled sandwiches together (1, 2, the sandwiches were called as demonstrated in figure 1 and listed below in table 1. The sandwiches were preheated at 280 °C for 7 min followed by ARB equals to 50%. The ARB experiments were executed, without lubricant, by using rolling machine. The rolling machine is a two-high rolling mill with 300 mm roll diameter, 600 mm width and a rolling speed of 0.40 m s⁻¹ it. The experimental steps are shown in figure 1. The most complex problem in the roll bonding step was small edge cracks. Tensile specimens were cut according to ASTM E8M shows in figure 2. Tensile testing was performed at room temperature with initial strain rate $1.1 \times 10^{-4} \text{ s}^{-1}$[12].

The different planes after ARB were identified by x-ray diffraction method (XRD, Bruker D8) using Cu Kα radiation ($\lambda = 1.5418 \text{ Å}$) with a scanning angle (2θ) of $10^\circ$–$100^\circ$ at a scan speed of 4 ($^\circ$)/min, a voltage of 40 KV and a current of 300 mA[13].

2.1. Microstructure preparation

Microstructure characterization studies were conducted on a metallographically polished specimens to investigate the morphology of grains. The metallographic samples after different cycles were cut with
The samples were ground consecutively with 400, 800, 1200 and 1500 grit Sic emery papers and polished using diamond paste. The microstructure of the polished samples was scrutinized by Field Emission Scanning Electron Microscope (FE-SEM) [14].

### 2.2. Tensile testing

#### 2.2.1. Mathematical design

For preparation of Al/Al₂O₃ composite using ARB technique, an experimental design, central composite design, was used to examine the impact of alumina content and number of cycles (ARB cycles) on the mechanical properties of aluminum/alumina composite. The design-matrix of different runs, 13 experiments, as well as the levels of each factor are illustrated in tables 2 and 3. Table 4 describes run, factors A, B and the UTS response.

The optimal conditions were estimated using a second order polynomial function by which a correlation between studied factors and response (UTS) was created. The general form of this equation is:

\[
E(y) = \beta_0 + \sum_{i=1}^{3} \beta_i X_i + \sum_{i=1}^{3} \sum_{j=1}^{3} \beta_{ij} X_i X_j
\]  

where y is the estimate of the response variable and Xi’s are the independent variables (Alumina content, and number of cycles) that are known for each experimental run. The parameters \(\beta_0, \beta_i, \text{ and } \beta_{ij}\) are the regression parameters. Software package, Design-Expert[13] was used for regression analysis of experimental data and to plot response surface. Analysis of variance (ANOVA) was used to assess the statistical parameters. The extent of fitting the experimental results to the polynomial model equation was stated by the determination coefficient, \(R^2\). F-test was used to estimate the significance of all terms in the polynomial equation within 95% confidence interval [15, 16].

### Table 1. Regime of the ARB process for production of Al/Alumina composite sheets.

| No. of cycles (Rolling) | Preheated Temperature(°C) | No. of Al-layers | Reduction in each cycle (%) | Total Reduction (%) | Total strain (ε) V.M |
|-------------------------|---------------------------|------------------|-----------------------------|---------------------|---------------------|
| 0                       | 300 for 5 min             | 2                | 67                          | 67                  | 12.45               |
|                         |                           |                  |                             |                     |                     |
| 1                       | 280 for 7 min             | 4                | 50                          | 50                  | 0.8                 |
| 2                       | 280 for 7 min             | 8                | 50                          | 75                  | 1.6                 |
| 3                       | 280 for 7 min             | 16               | 50                          | 87.5                | 2.4                 |
| 4                       | 280 for 7 min             | 32               | 50                          | 93.75               | 3.2                 |
| 5                       | 280 for 7 min             | 64               | 50                          | 96.87               | 4.0                 |
| 6                       | 280 for 7 min             | 128              | 50                          | 98.43               | 4.8                 |
| 7                       | 280 for 7 min             | 256              | 50                          | 99.21               | 5.6                 |
| 8                       | 280 for 7 min             | 512              | 50                          | 99.6                | 6.4                 |

![Tensile Testing](image)

**Figure 2.** Direction and dimensions of the UTS specimens prepared after ARB cycles (ASTM E8M).
3. Results and discussions

Figures 3, 4 and 5 show the composite longitudinal sections after cycles 1, 3 and 8. After cycle 1, that distributed layer of Al$_2$O$_3$ powder remains separate as detected in the longitudinal section. After the early passes, more discontinuous chains of alumina particles are observed between aluminum sheet layers, as shown in figure 4. For cycle 8, as it can be seen from figure 5, it is very difficult to recognize the number of Al layers in the micrograph, where boundaries of Al sheets layers disappeared. It can also be noticed that the thickness of alumina dense clusters decreases and consequently their size diminishes too. Figure 5 demonstrates that after the cycle 8, the uniformity of alumina clusters considerably increases. In other words, during the ARB process, the Al matrix flows between the alumina clusters, and consequently, the distance between the layers of Alumina particles increases. Cycle 8 (FE-SEM micrograph in figure 5) shows diffused interfaces with a good continuity. On the other hand, after the cycle 8, the porosities in the clusters are eliminated (well sound due good surface welding).

Table 2. Composite design has 3 levels and 2 variables (Al$_2$O$_3$ wt% and cycles No.).

| Column 1 Run No. | Factor A Alumina | Factor B Cycles |
|-----------------|------------------|----------------|
| 1               | −1               | 1              |
| 2               | 0                | −1             |
| 3               | 1                | 0              |
| 4               | 0                | 0              |
| 5               | −1               | −1             |
| 6               | −1               | 0              |
| 7               | 0                | 0              |
| 8               | 1                | −1             |
| 9               | 1                | 1              |
| 10              | 0                | 1              |
| 11              | 0                | 0              |
| 12              | 0                | 0              |
| 13              | 0                | 0              |

Table 3. Boundary conditions of accumulative roll bonded Al/Al$_2$O$_3$ composite according to design experts.

| Limits Variables | Minimum | Moderate | Maximum |
|------------------|---------|----------|---------|
| Alumina          | −1      | 3        | 5       |
| Cycles           | 2       | 5        | 8       |

Table 4. Factors and levels of experimental design using central composite method.

| Run No. | Factor A Alumina | Factor B Cycles | Response UTS, MPa |
|---------|------------------|-----------------|-------------------|
| 1       | 3                | 5               | 132               |
| 2       | 3                | 5               | 132               |
| 3       | 3                | 5               | 132               |
| 4       | 3                | 8               | 124               |
| 5       | 5                | 5               | 124.16            |
| 6       | 5                | 8               | 113.4             |
| 7       | 1                | 2               | 142               |
| 8       | 5                | 2               | 133               |
| 9       | 1                | 5               | 134.6             |
| 10      | 3                | 5               | 132               |
| 11      | 3                | 2               | 136               |
| 12      | 1                | 8               | 143.7             |
| 13      | 3                | 5               | 132               |
At early stages of ARB (Cycle 3), alumina layers are still apparent for 3wt% alumina sandwich. While in the late stages of ARB (Cycle 8), alumina layers disappeared, due to increase the aluminum layers. As it can be observed from figures 6 and 7, the behavior of Al/Al₂O₃ (3wt%) is similar as in Al/Al₂O₃ (1wt%). In addition, the thickness of alumina (3wt%) layers is larger than Al/Al₂O₃ (1wt%).
FE-SEM studies (figure 8) show thick interlayers of alumina particles between Al sheets after 7 ARB cycles. Alumina particles cannot deform plastically due to its ceramic nature; hence they create free space between them (horizontally or vertically). Adhesion between particles is not sufficient, so micro-cracks appear in these areas. On macroscale level, this is shown as weak bonding between the sheets, as demonstrated in figure 8.

3.1. Mechanism of dispersion of alumina particles
The FE-SEM micrographs at longitudinal sections of Al/1wt% Al₂O₃ composite were produced by ARB process in various cycles. It is observed that by increasing the number of ARB cycles, the laminate structure (the
aluminum sheets and powder layers) changed to a particle reinforced composite. There are several efficient mechanisms for changing the lamination structure to uniform dispersion of alumina particles in the Al matrix. At the initial cycles (early stages of ARB), during ARB the powder layers break up to small fragments and metal matrix of Al is extruded or squeezed through the fragments. Evidences of extrusion of the base metal through the powder layer fragments are detected in FE-SEM micrographs. As deformation of the harder agglomerates is considerably less than the matrix, the matrix flows past the fragments during the ARB, causing shear flow. The particles adjoining to the interface of fragments and matrix are less constrained than those in the interior of the fragments, and because of matrix shearing effect. They flow by tumbling along the fragment’s borders. By intensifying the strain (more ARB), the fragments continue to change their shape and their size as more particles are eliminated from the borders of the fragments (or Alumina agglomerates) and transferred to the end of the elongated clusters and/or agglomerations. FE-SEM shows elongated agglomeration with removed particles around it. By more deformations, the clusters or agglomerates show a preference orientation and gradually string themselves out along the rolling direction and finally, particles are regularly dispersed in the matrix. The fragments of the powder layer are completely disappeared at the last cycles of ARB process (late stages).

3.2. X-ray detection

3.2.1. Al/Al₂O₃ composites and ARB

Figure 9 shows the XRD of Al pure (annealed). The maximum peak is Plane (111) (closest plane). The second dominant plane is (200). The background is so smooth due to annealed structure. Figures 10, 11 and 12 show XRD plateau which explain synergistic effect of both ARB process and alumina particles content on the different peaks of aluminum/Al₂O₃ composite sheets. Generally, it is apparent that main plane (highest peak) of Plasticity for ARB sheet is (220) plane (dominant). Therefore, the elongation is sharply decreased than that of annealed one (main peak of aluminum annealed is (111)). Moreover, sheets containing 3 and 5wt% alumina, have dominant planes (111) and (200) after cycle 8 for 3wt% and after cycle 7 for 5wt% alumina. In addition, for
sheets containing 3wt% alumina after cycle 3 and cycle 0, the main planes (dominants) are (200) and (311), respectively. It is also clear that the background is rough due to high angle grain boundaries.

3.3. Mathematical modeling for Al/Al₂O₃ composite

The mathematical model boundary conditions are very important to avoid any irregularities of data response (UTS) and to choose the proper empirical equation which completely represents the whole data at different zones with very low errors. At first, cycle 0 data is excluded due to different preheating temperature (300 °C, 5 min), time and ARB ratio 67% while the rest of ARB experiments are carried out at 280 °C for 7 min having 50% reduction in thickness.

Figures 13 and 14 shows the contour behavior of alumina content (1, 3 and 5wt%) at number of cycles (ARB) on the ultimate tensile strength (UTS). At low, moderate or high cycle and 1wt% alumina, it is found that UTS slightly decreases till cycle 5 (142–136MPa) and then slightly increases again (136–143MPa).
From 2–3wt% alumina, it is clear that the effect of cycles after cycle 5 almost constant (133.6–128.6MPa). More than 3wt% alumina, on increasing the cycle, UTS gradually decreases.

The empirical equation that describes the mathematical relationship between alumina content and number of cycles on UTS is listed below. In general, it is obvious that both alumina and number of cycles decrease the UTS as indicated in the terms of alumina (91%) and number of cycles (667%). However, the number of cycles has more effect.

3.3.1. Final empirical formula in terms of actual factors (cycles and alumina) deduced by design expert software:

\[
\text{UTS} = 151.4586 - 0.91359 \times \text{Alumina} - 6.6745 \times \text{Cycles} \\
- 0.60912 \times \text{Alumina}^2 + 0.848075 \times \text{Cycles}^2 \\
+ 0.908333 \times \text{Alumina} \times \text{Cycles} + 0.127083 \\
+ 0.25583 \times \text{Alumina}^2 \times \text{Cycles}^2
\]

(2)

Figure 10. XRD of aluminum/1wt% alumina composites at different cycles.

Figure 11. XRD of aluminum/3wt% alumina composites at different cycles (ARB severity).

Figure 12. XRD of aluminum/5wt% alumina composites at different cycles (ARB severity).
The interaction between alumina content (wt%) and number of cycles is demonstrated in figure 15. Addition of 1 wt% alumina increases UTS than cycle 0 of composite Al/Al₂O₃. Furthermore, increasing the alumina content gradually decreases the UTS at low number of cycles. On the contrary, at high number of cycles, the UTS steeply decreases with increasing the alumina content. When the Alumina content increases, it agglomerates in the Aluminum matrix, weakens Al/Al₂O₃ composite and works as non-metallic inclusions according to Orowan concept [17]. Moreover, aluminum after severe plastic deformation exhibits to self-dynamic softening which decreases Al strength.

Figure 16 describes the relationship between the actual UTS from experiments and the predicted UTS from the empirical formula (deduced by Design Expert). This relationship can determine the error between actual and predicted values of UTS via correlation factor (R). It is found that the maximum error value is less than 2%.
4. Conclusions

1. Preheating ARB process can be used to produce high strength metal matrix composite.

2. FE-SEM microstructures disclose uniform distribution of alumina particles in the aluminum matrix, elongated in the rolling direction and good bonding of particles with the metal matrix at the interfaces.

3. X-ray diffraction of Pure Aluminum (annealed) exhibits main plane is (111) peak while for Al/Al₂O₃ composites, XRD show as that dominant plane changed to (111) and (200) peak due to ARB and Al₂O₃.

4. Tensile curves of Al/Alumina give maximum UTS of 169.5MPa and total elongation decreases to 0.08% (i.e. 99.7% decrease).

5. From the mathematical modeling, by adding Alumina content (1wt%) the UTS increases. While the UTS decreases with increasing the Alumina content (3 and 5wt%).

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References

[1] Fathy A, Abu-Oqail A and Wagih A 2018 Improved mechanical and wear properties of hybrid Al-Al2O3/GNPs electro-less coated Ni nanocomposite Cera. Inter. 44 22135–45
[2] Liu T, Wang Q, Sui Y, Wang Q and Ding W 2016 An investigation into interface formation and mechanical properties of aluminum–copper bimetal by squeeze casting Mater. Des. 89 1137–46
[3] Cui C, Schulz A, Schimanski K and Zoch H-W 2009 Spray forming of hypereutectic Al–Si alloys J. Mater. Process. Technol. 209 5220–8
[4] Jamaati R, Toroghinejad M R, Dutkiewicz J and Szpunar J A 2012 Investigation of nanostructured Al/Al2O3 composite produced by accumulative roll bonding process Mater. Des. 35 37–42
[5] Ramu G and Bauri R 2009 Effect of equal channel angular pressing (ECAP) on microstructure and properties of Al–SiCp composites Mater. Des. 30 3554–9
[6] Kapoor R, Sarkar A, Yogi R, Shekawat S K, Samajdar I and Chakravartty J K 2013 Softening of Al during multi-axial forging in a channel die Mater. Sci. Eng. A 560 404–12
[7] Lee S-H, Sakai T, Saito Y, Utsunomiya H and Tsuji N 1999 Strengthening of sheath-rolled aluminum based MMC by the ARB process Mater. Trans., JIM 40 1422–8
[8] Reihanian M, Shahmansouri M J and Khorasanian M 2015 High strength Al with uniformly distributed Al2O3 fragments fabricated by accumulative roll bonding and plasma electrolytic oxidation Mater. Sci. Eng. A 640 195–9
[9] Jamaati R, Toroghinejad M R, Hoseini M and Sazpunar J A 2012 Development of texture during ARB in metal matrix composite Mater. Sci. Technol. 28 406–10
[10] Ghalabbandi S M, Malaki M and Gupta M 2019 Accumulative roll bonding—a review Appl. Sci. 9 3627
[11] Alizadeh M 2012 Effects of temperature and B4C content on the bonding properties of roll-bonded aluminum strips J. Mater. Sci. 47 4689–95
[12] Hou Z, Gutierrez M, Ma S, Almrabat A and Yang C 2019 Mechanical behavior of shale at different strain rates Rock Mech. Rock Eng. 52 1–14
[13] Il Langford J and Wilson A J C 1978 Scherrer after sixty years: a survey and some new results in the determination of crystallite size J. Appl. Crystallogr. 11 102–13
[14] Fathy A and El-Kady O 2013 Thermal expansion and thermal conductivity characteristics of Cu–Al2O3 nanocomposites Mater. Des. 46 355–9
[15] Abu-Oqail M and Moataz H A 2019 Enhancement ductility of friction stir welded aluminum blanks AA2024 via adding interlayer strip width of AA7075 Materials Research Express 6 6–11
[16] Serio L, Palumbo D, De Filippis L, Galietti U and Ludovico A 2016 Effect of friction stir process parameters on the mechanical and thermal behavior of 5754–H111 aluminum plates Materials (Basel). 9 122
[17] Humphreys F J M H 2013 Recrystallization and Related Annealing Phenomena 53